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Office of Advanced Automotive Technologies
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FY 1999

Combustion and Emission Control Progress Report

Energy Efficiency and Renewable Energy
Office of Transportation Technologies
Office of Advanced Automotive Technologies
Energy Conversion Team

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I. INTRODUCTION

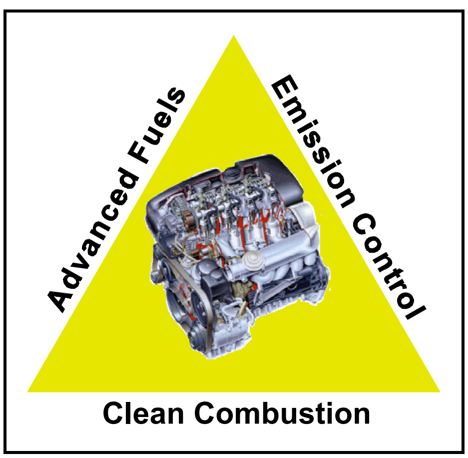
Developing Advanced Combustion and Emission Control Technologies

Kenneth Howden, Program Manager

On behalf of the Department of Energy's Office of Advanced Automotive Technologies (OAAT), I am pleased to introduce the Fiscal Year (FY) 1999 Annual Progress Report for the Advanced Combustion and Emission Control (Aftertreatment) Research and Development (R&D) Program. This introduction serves to briefly outline the nature, progress, and future directions of the Advanced Combustion and Emission Control R&D Program. Together with DOE National Laboratories and in partnership with private industry and universities across the United States, OAAT engages in high risk research and development that provides enabling technology for fuel efficient and environmentally-friendly light duty vehicles.

PNGV: Technologies that enable 80 mpg fuel economy in passenger cars with low emissions

The Advanced Combustion and Emission Control Research and Development (R&D) Program supports the Partnership for a New Generation of Vehicles (PNGV), a government-industry partnership striving to develop, by 2004, a mid-sized passenger vehicle capable of achieving 80 miles per gallon while adhering to future emissions standards and maintaining such attributes as affordability, performance, safety, and comfort. Following a rigorous process of technology down-selection, PNGV has targeted compression-ignition direct-injection (CIDI) engines, an advanced version of the commonly known diesel engine, as one of the promising technologies for achieving the 80 miles per gallon fuel economy in a light-weight hybrid vehicle. Today's CIDI engines achieve impressive thermal efficiency, however, in order to meet proposed future emissions standards, advancements in clean combustion, emission control technology and diesel fuels are necessary.



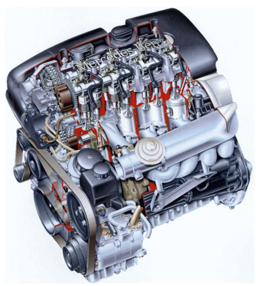
The Advanced Diesel Engine is a targeted PNGV Powerplant Technology

The Advanced Combustion and Emission Control R&D Program explores the fundamentals of combustion, how emissions are formed, and advanced methods for treating those emissions. Testing and modeling are also important elements of the program and enable us to evaluate potential technology and validate technology selection and direction. By working at the forefront of these new technologies, we hope to enhance the knowledge base that can be used by automotive partners and suppliers (engine manufacturers, catalyst companies, etc.) to develop advanced automotive systems.

Our program, which is cost-shared between industry and government, is sharply focused on improving combustion processes and emission control technologies. Progress in these areas will help overcome the critical technical barriers associated with the emission challenges of highly efficient CIDI engines, namely, high levels of nitrogen oxides (NO_X) and particulate matter (PM) in the exhaust. OAAT is also pursuing developments in automotive fuels through the Advanced Petroleum-Based Fuels Program, which is described in a separate report.

Technical Leadership

Lean Burn Catalyst for NO_X Reduction



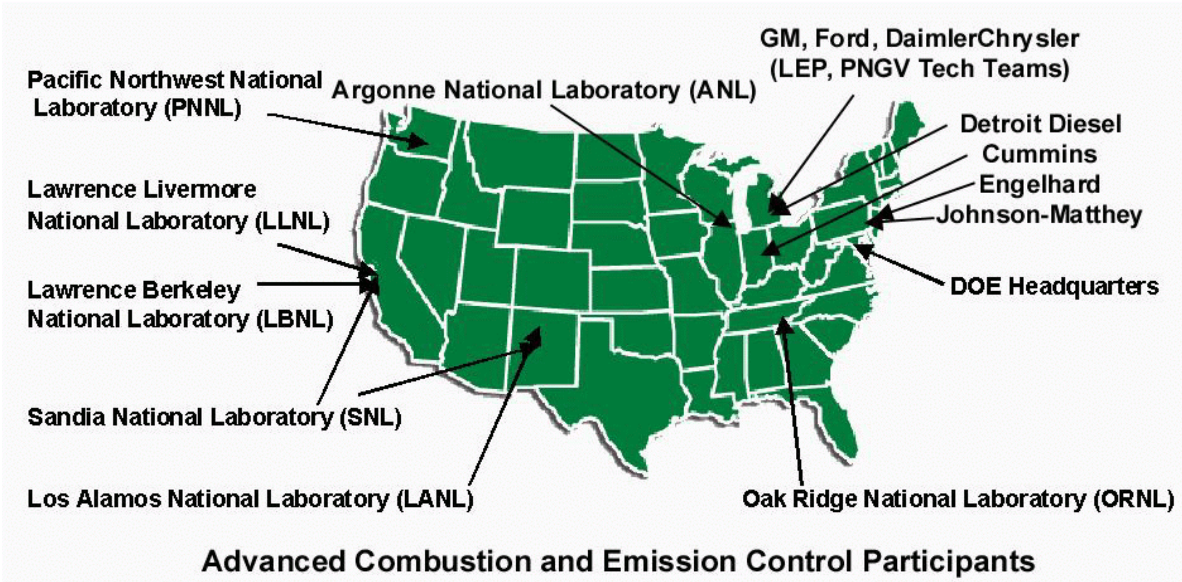
Advanced Diesel Engine

High Pressure Common Rail Fuel Injection

By providing strong technical direction and program and financial management, DOE has taken a leadership role in the development of technology to reduce exhaust emissions of highly efficient diesel engines without compromising cost, performance or efficiency. While not directly funding the U.S. automakers, DOE could not provide this leadership without General Motors, DaimlerChrysler and Ford participating through PNGV technical teams and the Low Emission Partnership (LEP). The technology developed under our program will help the automakers overcome a critical barrier to producing light-duty vehicles with up to three times the fuel economy of today's conventional vehicles while meeting strict emissions standards established by the EPA and the California Air Resources Board (CARB).

The unique scientific capabilities and facilities of the National Laboratories offer us the opportunity to target those technical areas of highest priority and those that require high-risk R&D that is seldom conducted independently by industry. We work with industry and through the PNGV technical teams to ensure that National Laboratory R&D activities fulfill industry's needs for basic scientific understanding and development of new technologies, devices, materials, and processes.

In response to recommendations made by the National Research Council encouraging much greater participation by industry in the Advanced Combustion and Emission Control Program, we have expanded our work plan to include two new projects with leading U.S. diesel engine manufacturers and emission control manufacturers. The expanded work plan has led to an increase in program resources, which grew from $7.6 million in FY 1998 to $12.8 million in FY 1999. In FY 1999, our program awarded competitive contracts to Detroit Diesel Corporation, working in partnership with Johnson-Matthey, and Cummins Engine Company, working in partnership with Engelhard Corporation, to develop advanced emission control systems for both passenger cars and light trucks. Each partnership will pursue several combinations of emission control devices and then down-select to a system which is optimized for a small-displacement (under two liters) 3-cylinder PNGV CIDI engine. They will then scale up to a second system designed for a new light-truck, four-liter V6 CIDI engine.



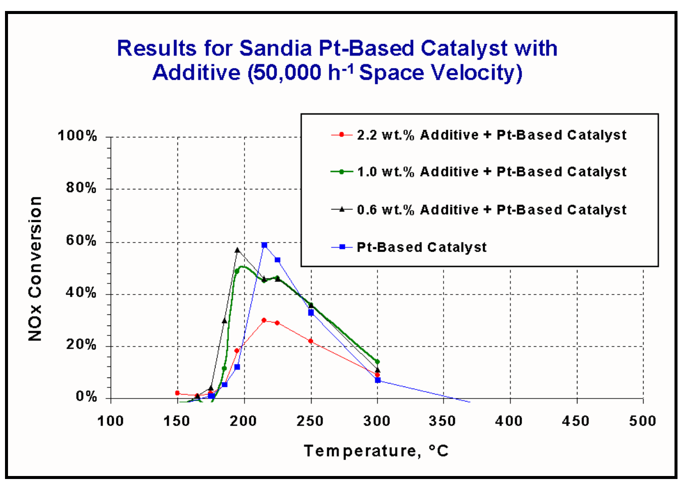
Challenges

As indicated above, the number one technical barrier for CIDI engines is reducing and controlling unwanted NO_X and PM in the exhaust. In order for CIDI engine technology to be successfully integrated into a PNGV vehicle, it must effectively decrease emissions without jeopardizing fuel economy. To meet proposed EPA Tier 2 standards, NO_X emissions and particulate emissions will have to be reduced by over 85 percent.

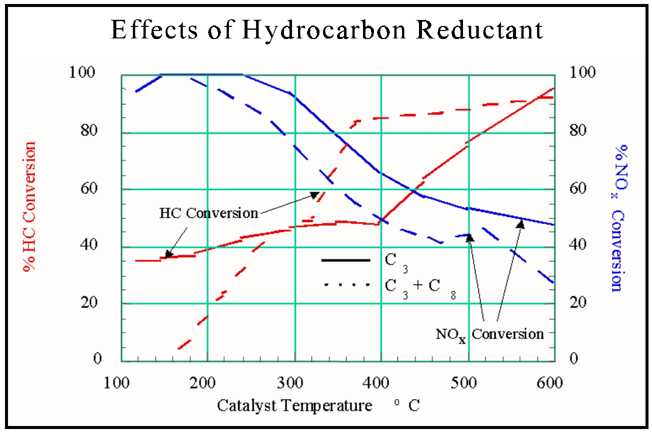
OAAT programs have been successful in meeting many of the original milestones established by PNGV. In response to the proposed EPA Tier 2 standards, OAAT and its industry partners are setting new research targets for CIDI emissions control systems. We believe that further advancement in CIDI engines, emission control devices and diesel fuel formulations may result in emissions low enough to meet these proposed federal standards. DOE, national laboratories and industry partners recognize the capital, resource, and technical investment necessary and are poised to make the necessary investments to tackle these new challenges.

Accomplishments

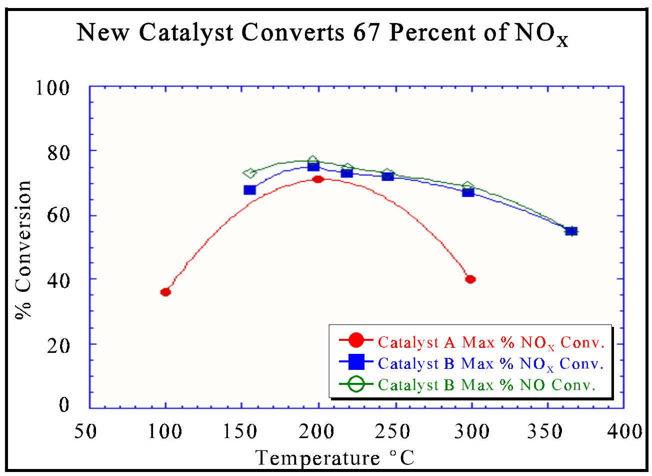
In FY 1999, we saw impressive results in new catalyst technology to reduce NO_X emissions, novel methods to reduce PM emissions, improvements in combustion processes using exhaust gas recirculation (EGR), and more detailed characterization of combustion processes.



Sandia National Laboratory (SNL) developed a new catalyst material that improves catalyst performance by lowering light-off temperature and widening the temperature window of appreciable NO_X reduction. In FY 1999, SNL also completed the first phase of technology transfer of hydrous metal oxide-supported catalyst technology to the LEP. A process was defined to enable transfer of catalyst materials synthesis procedures to participants, and signed nondisclosure agreements are in place with four major catalyst suppliers. Although the technologies developed by SNL themselves will not meet the stringent EPA standards proposed for 2004, they do contribute substantially to the catalytic materials knowledge base and lay the groundwork for future cooperation and technology transfer.



Los Alamos National Laboratory (LANL) developed a new zeolite-supported catalyst that achieves high NO_X conversion rates. LANL has discovered microporous catalysts that contain single transition element ions that convert NO_X in simulated diesel exhaust with high efficiency on laboratory samples. Not only do these catalysts have high rates of NO_X removal, but they perform this catalysis over a broad temperature range, converting more than 95% of the NO_X between 250 - 500 degrees Centigrade.



Pacific Northwest National Laboratory (PNNL) demonstrated high NO_X conversion rates for a plasma-catalyst in tests using simulated exhaust with sulfur added and realistic space velocities. Preliminary testing is now being conducted on actual full-scale engine exhaust flow. The plasma reactor and its power supply are being continuously refined to increase conversion efficiency and reduce power consumption.

Shifting focus from port-fuel-injected gasoline engines to CIDI engines has required a corresponding shift in emission control technology from oxidation catalysts to lean NO_X catalysts. The LEP is an essential forum for automotive industry researchers and the national laboratories, providing a systems-level perspective for setting emission control performance goals and linking major suppliers to the program. We are proud of the strong working relationship between DOE, LEP, equipment suppliers, and national laboratories and look forward to further strengthening these partnerships in the coming year.

Future Directions

Our new program directions for FY 2000 build upon recent advances in technology development and begin to move some of the more promising new technologies out of the laboratory and over to industry for testing and implementation. Several developments will build upon and enhance the work completed this year:

- Availability of Prototype Engines: To date, most testing has been conducted using state-of-the-art CIDI engines. In order to meet the proposed Tier 2 standards and to address concerns about ultra-fine particulate and toxic emissions, significant advances in engine design to reduce engine-out emissions are needed. Next year, prototype engines should become available which will provide a much more realistic baseline for evaluating emission control device performance and the effects of fuel reformulations.

- Development of New Tools to Evaluate PM: As emission standards become more stringent, accurate measurement of PM becomes more critical to identify their origins and devise methodologies for further reductions. For instance, it is believed that transients are responsible for much of the PM produced but it is not possible now to accurately allocate PM to transient modes. The most promising technology for measuring transient PM emissions appears to be laser light scattering. Using this technology, real-time measurement of PM volume fraction, primary particle size, and volume-equivalent-sphere diameter is believed to be possible and could contribute significantly to understanding PM production.

- Fundamental Combustion Research: Understanding the formation of PM and NO_X holds the key to devising technology for controlling emissions. Formation of PM and NO_X will be addressed on three separate fronts: 1) laser diagnostics that provide visual images of PM and NO_X formation; 2) chemical kinetics that predict the combustion reactions that form PM and NO_X; and 3) advancements in computational fluid dynamics which are used to model air and fuel flow within the engine.

- Emission Control Device Research, Development & Testing: Previous work has identified several promising technologies for emission control devices to reduce PM and NO_X emissions. The focus for the next year will be on how to improve the efficiency and durability of these devices. A potential ruling from EPA on the sulfur level of diesel fuel will facilitate deciding which technologies are most promising and accelerate development of practical designs. Some of the specific activities that will be completed include:

  - Full-scale prototypes of non-thermal plasma devices and self-regenerating particulate filters will be tested using CIDI engine exhaust streams.
  - OEMs and catalyst suppliers will test new LANL-developed microporous catalyst materials that promise much higher NO_X conversion over a wide exhaust temperature range.
  - Selected PM and NO_X control devices will be tested for durability with the prototype DDC CIDI engines being developed.
  - Though an established technology for stationary applications, selective catalytic reduction (SCR) which employs urea injection and a special catalyst to convert NO_X is being considered by the LEP for testing in the coming year.
  - Initiation of two new cooperative agreements with U.S. diesel engine manufacturers and catalyst industry partners will enable the integration of advanced devices into emission control systems capable of meeting future stringent standards for both passenger car and light truck applications.

Honors and Special Recognitions

The figure below contains a selected group of patents filed by and/or issued to National Laboratories in FY 1999 which reflect some of the exciting work conducted at the Labs under the auspices of the Advanced Combustion and Emission Control R&D Program.

Selected Patents Filed and/or Issued in FY 1999

- **ORNL/FORD**: Method of Controlling Cyclic Variation in Engine Combustion
- **SNL**: Nitrogen Oxide Absorbing Material; Material and System for Catalytic Reduction of Nitrogen Oxide in an Exhaust Stream of Combustion Process
- **PNNL**: Method and Apparatus for Exhaust Gas; Exhaust System and Emissions Storage Device and Plasma Reactor; Catalytic Plasma Reduction of NO_X
- **ANL**: Method and Apparatus for Reducing Particulates and NO_X Emissions from Diesel Engines Utilizing Enriched Combustion Air; Method to Reduce Diesel Engine Emissions Parts 1&2; Method and Apparatus for Reducing Cold-Phase Emissions by Utilizing Oxygen-Enriched Intake Air; Variable Oxygen/Nitrogen-Enriched Intake Air Systems for Internal Combustion Engine Applications
- **LANL**: Microporous Catalysts for NO_X Conversion

John E. Dec, Dennis L. Siebers and Peter O. Witze (SNL) - Honored as SAE Fellows for in-cylinder fluid motion and combustion phenomena research at 1999 SAE International Congress and Exposition in Detroit.

Lockheed Martin Energy Research Corporation honored ORNL scientists John M. Storey and Karren More for their cutting edge research to reduce emissions and mitigate potential health effects from diesel engines.

A team from Sandia National Laboratory, including Tim Gardner, Steve Lockwood, Linda McLaughlin, and Ron Sandoval, garnered the 1999 National Laboratory CIDI R&D Award for their outstanding achievement in research and development of lean NO_X catalysts for diesel engines and engine exhaust emission control.

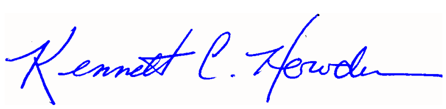
Conclusion

The remainder of this report highlights progress achieved during FY 1999 under the Advanced Combustion and Emission Control R&D Program. The following 24 abstracts of industry and National Lab projects provide an overview of the exciting work being conducted to tackle tough technical challenges associated with CIDI engines, including fuel injection, exhaust gas recirculation, fuel mixing, combustion processes, and catalytic exhaust treatment devices for controlling emissions.

R&D Magazine awarded ANL an R&D 100 Award for "Clean Diesel Technology: Simultaneous reduction of NO_X and Particulates from diesel engines by three-way optimization of oxygen-purity in the intake air, fueling rate and timing."

PNNL's Mary Lou Balmer was awarded the Presidential Early Career Award for plasma-activated catalysis work

We are encouraged by the technical progress realized under this dynamic program in FY 1999. We also remain cognizant of the significant technical hurdles that lie ahead, especially those presented for CIDI engines by the proposed EPA Tier 2 emission standards that are scheduled to go into effect beginning in 2004. In FY 2000, we look forward to working with our industrial and scientific partners to overcome many of the barriers that still stand in the way of delivering advanced automotive technology.



Kenneth Howden, Program Manager
Combustion and Emission Control R&D Program
Energy Conversion Team
Office of Advanced Automotive Technology (OAAT)

II. EMISSION CONTROL/SUB-SYSTEMS DEVELOPMENT

II.A. Application of Advanced Emission Control Sub-systems to State-of-the-Art Diesel Engines

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Subcontractors
Johnson Matthey, Wayne, PA
Michigan Technical University, Houghton, MI

Objectives

- Demonstrate technology which will achieve tailpipe emissions levels that meet PNGV Year 2004 emissions targets in a hybrid-electric vehicle (HEV)
- Demonstrate scalability of the technology to a light truck/sport-utility-vehicle-sized engine
- Meet specified targets for engine-out emissions, efficiency, power density and noise, durability, production cost, and aftertreatment volume and weight.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 5, Barriers A,B,C

Approach

- Model emission control sub-systems: Johnson Matthey for NO_X control; Michigan Technical University for particulate matter (PM) control.
- Generate real-time speciated emissions data as input for models.
- Integrate engine and emission control sub-system models.
- Select and evaluate best engine/emission control sub-systems.
- Conduct engine dynamometer durability testing.
- Scale emission control sub-systems for SUV engine.
- Demonstrate SUV engine in a vehicle.

Accomplishments

- This project was awarded in late FY 99.

Future Directions

- Initiate work following contract finalization.
- Put subcontracts into place.
- Deliver inline 3-cylinder and V6 CIDI engines with associated emission control sub-systems.

Background

DDC was recently selected for participation in a DOE project entitled "Research and Development for Compression-Ignition Direct-Injection Engines (CIDI) and Aftertreatment Subsystems." This 30-month program calls for development of aftertreatment technologies on a PNGV-sized engine, and to demonstrate scalability of the technologies to a light truck/SUV-sized engine. DDC is in the unique position of being the only engine manufacturer developing engines for both the PNGV and SUV programs.

Objectives

This project will demonstrate technology which will achieve tailpipe emissions levels that meet PNGV Year 2004 emission targets in an HEV vehicle. This will be done using a 1.46L 3-cylinder engine which DDC has developed and supplied to DaimlerChrysler (DC) under the PNGV program. This engine is illustrated in Figure 1. The specific targets are 0.2 g/mi NOx and 0.01 g/mi PM at HEV vehicle weight and engine power requirement (based upon DC HEV). The project will also demonstrate scalability of the technology to a light truck/SUV-sized engine of 3.0-4.0L displacement, such as the 4.0L V6 DELTA engine (shown in Figure 2) developed in cooperation with the DOE Office of Heavy Vehicle Technologies. The engines will meet specified targets for engine-out emissions; efficiency; power density and noise; durability (both engine and aftertreatment); production cost (both engine and aftertreatment); and

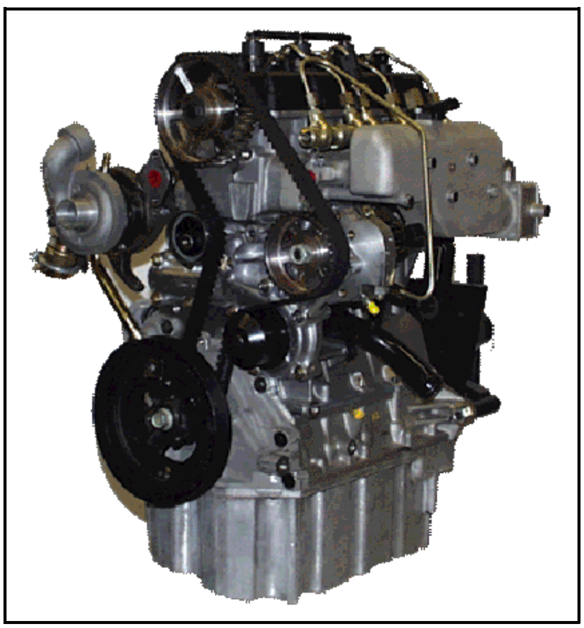

Figure 1. 3-Cylinder CIDI HEV Engine

Figure 2. 6-Cylinder CIDI SUV Engine (DELTA V6)

aftertreatment volume and weight.

Approach

In this project, aftertreatment subsystem models will be developed that effectively predict aftertreatment performance under a variety of input conditions. Johnson Matthey has been selected as subcontractor to conduct the NOx aftertreatment modeling, and Michigan Technological University has been selected to perform the PM aftertreatment modeling. DDC will generate and provide real time speciated emissions stream data for use as inputs to the models. Based on the results of the predictive models, the best combination(s) of NOx and PM aftertreatment will be selected and further developed. DDC will integrate the aftertreatment models with engine models into an overall predictive model for the integrated systems. Based on the model predictions, engine emission reduction technologies that pair with the selected aftertreatment technologies will be developed to provide the best overall vehicle tailpipe emissions. Final selection and evaluation of the best engine/aftertreatment technology combination(s) will be done using mechanical drive Dodge Neon vehicles. Durability of the selected combination will be conducted using engine dynamometer testing. A schematic of the development process is shown in Figure 3.

After selection of the aftertreatment for the HEV-sized engine, modeling will be used to scale the aftertreatment for the V6 SUV engine. Combined with scaled engine technologies, the system will be developed on the V6 engine in the laboratory and then demonstrated in a vehicle which has been retrofitted with the engine.

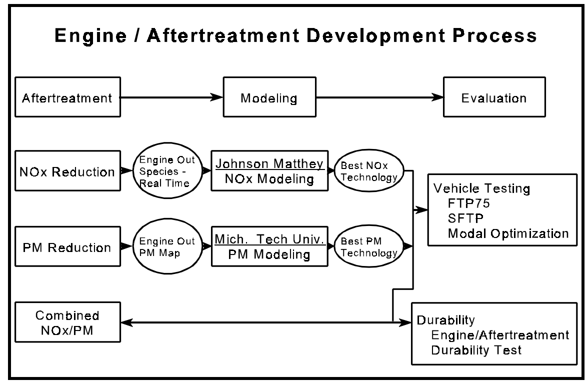

Figure 3. Engine/Aftertreatment Development Flow Chart

Deliverables

At the end of the 30-month project, DDC will deliver to DOE both a 3-cylinder HEV engine and a 4.0L V6 DELTA engine along with their corresponding aftertreatment subsystems. The delivered technologies will demonstrate the lowest possible tailpipe emissions in light duty vehicles believed to be achievable utilizing technologies that can be cost effectively produced. With further development, it is believed that these technologies also form the basis for meeting stretch emissions goals that are consistent with proposed Tier 2 standards.

List of Acronyms

CIDI	Compression Ignition Direct Injection
DC	DaimlerChrysler
DDC	Detroit Diesel Corporation
HEV	Hybrid Electric Vehicle
PM	Particulate Matter
PNGV	Partnership for a New Generation of Vehicles
SUV	Sport Utility Vehicle

II.B. Technologies for NO_X and Particulate Matter Reductions for Advanced Diesel Engines

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Subcontractors
Englehard, Newark, NJ
FEV, Detroit, MI
Richardson Electronics, Chicago, IL
Plasma Technics, Racine, WI
ECI/Lo Tec, San Diego, CA

Objectives

- Develop aftertreatment technologies applicable for LDV and LDT engines ranging from 55 kW to 200 kW.
- Deliver an optimized aftertreatment sub-system for a 55 kW engine for PNGV applications to achieve the project goals of less than 0.20 g/mile oxides of nitrogen (NO_X) and 0.010 g/mile particulate matter (PM) with engine-out emissions of 1.4 g/kW-hr NO_X and 0.15 g/kW-hr PM. Only those technologies which have a reasonable chance (with further development beyond the scope of this program) of meeting proposed EPA Tier 2 regulations of 0.07 g/mile NO_X and 0.01 g/mile PM will be pursued.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 5, Barriers A,B,C

Approach

- Under this 30-month program, various aftertreatment technologies including non-thermal plasmas, NO_X adsorbers, and lean NO_X catalysts, in conjunction with active reductant injection will be investigated to address NO_X emissions. The areas of development include catalyst formulation for high NO_X conversion over a wider catalyst/exhaust gas temperature range, catalyst structure for increased exhaust gas residence time on active catalyst sites, and an understanding of the various factors that cause deactivation of the catalyst. Fuel reformulation concepts and diesel fuel based onboard hydrocarbon cracking strategies will be investigated to increase the activity of the catalyst systems. Even with the availability of 30 ppm sulfur fuels, the development of a sulfur management scheme is critical to prevent catalyst poisoning and deactivation. The application of a sulfur trap that can be regenerated or periodically replaced will be explored.

- PM emissions will be addressed by developing a catalyzed soot filter, a microwave heated particulate filter, or a combination of catalyzed soot filters with supplemental microwave heating in parallel with a NO_X reduction system. Soot filter catalysts have been successfully formulated for heavy-duty applications with passive regeneration. However, with the lower exhaust temperatures anticipated for PNGV applications, an active regeneration scheme with supplemental heating will be investigated.
- Finally, the improved aftertreatment components will be integrated and configured optimally in a system developed for a PNGV application. This system will then be calibrated and tested in a controlled environment on a PNGV sized engine.

Accomplishments

- This project was awarded in late FY 99.
- A meeting was conducted with the Oak Ridge National Laboratory (ORNL) to discuss verification testing of the PNGV exhaust aftertreatment subsystem. Test procedure, hardware requirements, technical support requirements, and schedule were all discussed.
- Plasma Assisted Catalytic Reduction Aftertreatment System—The majority of the work completed on the PACR engine was centered around the development of the 12 steady state modes to be used on the ISB engine when developing the PACR aftertreatment system. An Assessment of Critical Characteristics vs. Options matrix has been constructed for the PACR EAS system. This matrix was developed using a joint working session between Cummins and Engelhard. Several action items have been identified from the matrix that will be followed up by Engelhard.
- NO_X Adsorber Catalyst—A framework for a controls model has been established and an idea of what data will be required has been discussed. Engelhard is currently collecting reactor data to support this model and Cummins is developing engine tests to verify the reactor results.
- Exhaust Aftertreatment Subsystem Modeling—The transient thermal model for the take-down pipe was extended to the monolithic catalyst. The model included heat convection between the exhaust gas and catalyst surface, thermal inertia of the catalyst, and heat convection / radiation to the ambient. An air gap insulation model was added to simulate the effect of reducing heat loss to the ambient. Preliminary comparison with FTP-75 test results using measured engine-out temperature as input showed generally good agreement in the predicted catalyst-out temperature. The model clearly showed a strong thermal damping effect of the catalyst, as the temperature variations at the exit of the catalyst were considerably reduced.
- FEV was consulted about the necessary personnel and hardware support of DIATA engines for EAS validation testing and final configuration optimization. FEV has requested a letter from Ford authorizing them to make information/support for the DIATA engines available to Cummins Engine Co.

Future Directions

- Plans are in place for the project kick-off meeting which is targeted for the month of October, in anticipation of contract finalization.
- Plasma Assisted Catalytic Reduction Aftertreatment System—The CRT upstream of the Gamma-Alumina catalyst will be tested and compared to the 12 mode DOE baseline using Gamma - Alumina alone.
- Lean NO_X Catalyst—The reactor map data will be verified. Several catalyst configurations will be

studied, including different orders of precious metals & base metals, different injection locations, injections between bricks, and series versus parallel.

- Exhaust Aftertreatment Subsystem Modeling—Work will begin to study the chemical reactions with the lean NO_X catalyst to understand the various competing mechanisms involved. The heat generated by HC oxidation, as predicted by the chemical reaction model, will become inputs to the simulation and alter the catalyst thermal response.

Overview

The proposed EPA Tier 2 NO_X standards will be more stringent than the PNGV target. As a result, the need to bring the NO_X goals for the cooperative agreements into harmony with the anticipated EPA regulations required that modification be made to the program targets. Cummins has agreed to modify the existing targets to include an interim NO_X target of less than 0.2 g/mile and a stipulation that only those technologies which have a reasonable chance (with further development beyond the scope of this program) of meeting EPA anticipated regulations of 0.07 NO_X g/mile and 0.01 PM g/mile be pursued. The program's primary focus will be on aftertreatment subsystem NO_X and PM conversion efficiency, although the higher conversion efficiency required may result in an increase in the cost, size, and weight of the aftertreatment subsystem.

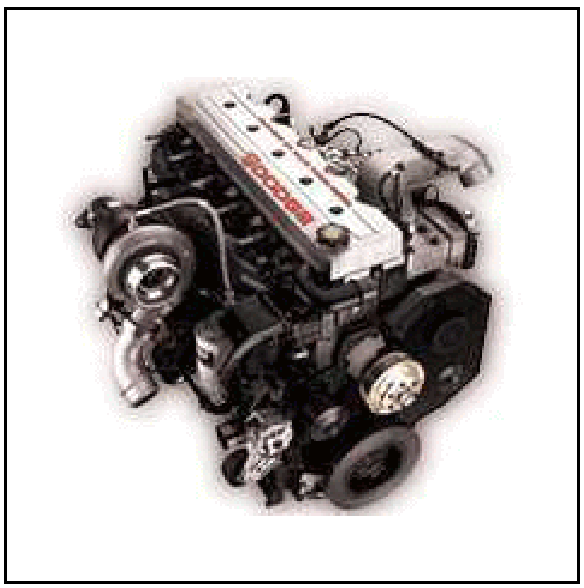

Figure 1. Cummins ISB Engine

Figure 1. Ford DIATA Engine

The key objective of this project, as stated previously, is to develop the generic aftertreatment technologies applicable for LDV and LDT engines ranging from 55 kW to 200 kW. This will involve engines with displacement ranging from 1.2 liters to 6.0 liters. A fundamental and 'displacement-size' transparent understanding will be required. Cummins' initial results indicate that the LDV and LDT exhaust operating characteristics can be simulated with the Cummins ISB mule engines (see Figure 1). Therefore, most of our aftertreatment subsystem screening and fundamental understanding will be conducted on the ISB mule engines. In addition, parallel performance validation and final system optimization will be conducted on a PNGV sized engine approved by DOE. Cummins is planning to use the DIATA engines, developed under the Ford Hybrid Propulsion

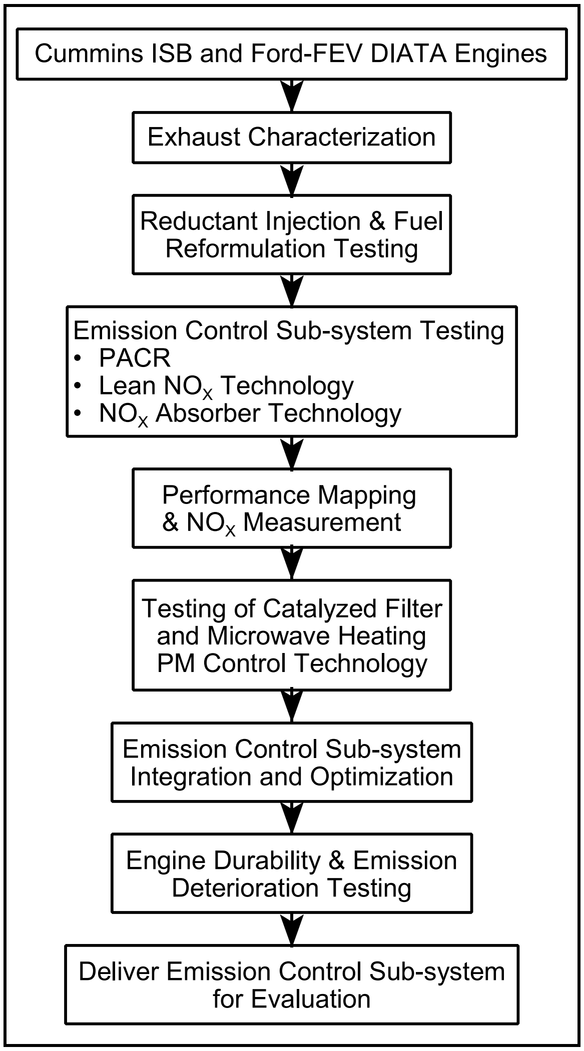

Figure 3. Flowchart of Project Activities

Systems Development Program, as the focus of this project (see Figure 2). Three DIATA engines will be delivered to Cummins by the end of October 1999. The engines need to be optimized and capable of twelve-mode steady state composite and/or simulated FTP-75 transient cycle operation depending on the final evaluation requirements at ORNL. Arrangements have been made to obtain technical support in a way that will ensure that intellectual property is safeguarded for all affected parties. Cummins will utilize DIATA engine data and PNGV target vehicle/engine-out emissions to determine the input specifications for modeling and design optimization. This effort is required to provide evidence through modeling that the results recorded with the engine dynamometer test can be expected to meet the vehicle emission requirement. Figure 3 is a detailed flowchart of the activities described above.

Close cooperation and interaction with the PNGV 4SDI technical team members is crucial to the overall success of the program. Therefore, the 4SDI Technical Team members will participate in the Cummins kick-off meeting, as well as be involved with the semi-annual reviews, during which progress against PNGV targets will be evaluated.

Publications/Presentations

Roland M. Gravel, "Looking Toward Tier II - A Program to Address Diesel Emissions," being prepared for SAE 2000 Future Car Congress, Arlington, VA, April 2000

List of Acronyms

CRT	Continuously Regenerating Trap
DIATA	Direct Injection, Aluminum, Through-Bolt Assembly
EAS	Exhaust Aftertreatment Sub-System
ISB	Interact System B (a fully electronic, 24 valve inline 6-cylinder B-family engine)
ORNL	Oak Ridge National Laboratory
PACR	Plasma Assisted Catalytic Reduction System
PNGV	Partnership for a New Generation of Vehicles
4SDI	4-Stroke Direct Injection

III. IN-CYLINDER COMBUSTION STUDIES, ADVANCED COMBUSTION RESEARCH, AND DIAGNOSTICS

III.A. Advanced Diesel Combustion R&D

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Subcontractors
University of Wisconsin Engine Research Center (UW ERC), Madison, WI
Wayne State University (WSU), Detroit, MI

Objective

- To provide the physical understanding of in-cylinder combustion processes needed to meet the efficiency and emissions standards of the PNGV vehicle.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 4, Barriers A,B

Approach

- Obtain measurements which clarify the fundamental in-cylinder physical processes important to high-speed, direct-injection (HSDI) diesel combustion through use of optical diagnostic techniques in a geometrically-realistic optical engine.
- Correlate in-cylinder processes observed in the optical engine to tailpipe emissions and traditional, pressure-based performance analysis in a conventional single-cylinder test engine with geometry and operating conditions as close as possible to the optical engine.
- Incorporate fundamental physical processes into detailed computer models of engine combustion; validate the output of these models against tradition test engine emissions and performance.

Accomplishments

- Established three industrially relevant key operating points for detailed testing, which are representative of approximately 80% of the FTP cycle.
- Characterized optical engine combustion performance at key operating points, and validated against comparable production engine data.
- Identified locations of ignition and early combustion at key operating points using simultaneous,

2-camera imaging of natural flame chemiluminescence.
- Examined locations of soot formation and spatial distributions in-cylinder using 2-camera imaging of soot luminosity.
- Investigated the effects of pilot injection and swirl ratios between 1.5 and 4.0 on ignition locations, soot formation timing and spatial location, combustion performance and combustion noise.
- Completed laboratory set-up for traditional test engine, including sub-systems for control of engine air, lubrication, EGR, cooling, and emissions measurement (WSU).
- Commenced characterization of test engine operation at key operating points and comparison with optical engine (WSU).
- Completed initial full-cycle computer simulations, assessing mesh integrity and appropriate methods of assigning initial conditions for computations (UW ERC).
- Validated appropriate thermodynamic state at TDC for optical engine by computing cylinder pressures and bulk gas temperatures and comparing with computations of the traditional test engine (UW ERC).

Future Directions

- Perform additional studies of ignition and soot formation to complete the matrix of test conditions, including different injector tip geometries.
- Measure and validate computer modeling of the velocity field in the combustion bowl at the start of injection for swirl ratios between 1.5 and 4.0.
- Investigate the effect of variable swirl on the liquid penetration length of the fuel jets.
- Characterize and compare the combustion performance of the traditional test engine to the optical engine and comparable production engines; characterize the test engine emissions performance with varying EGR levels (WSU).
- Continue physical sub-model development and computational support of experimental work; commence detailed comparison of computed results with data obtained in the optical and traditional test engines (UW ERC).

Introduction

The physical processes important in HSDI diesel combustion are more complicated than those prevailing in heavy-duty size class engines due to the strong, swirling fluid motion and interaction of the fuel sprays and combusting jets with the walls of the combustion chamber. The deliverable of this project is to develop a physical understanding of how the combustion and emissions formation processes are influenced by these complicating factors, and to incorporate this understanding into computational models that can be used as engine design tools. Several aspects of the combustion process are expected

Table 1. Specifications of the HSDI Engine

Geometry	4-Valve, Central Injector
Bore	7.95 cm
Stroke	8.50 cm
Speed Range	0-4,000 rpm
Swirl Ratio	$1.5 < R_s < 4.0$
Flexible FIE	Fiat/Bosch HEUI HI-90
Target Performance	18 bar IMEP; 45 kW/liter

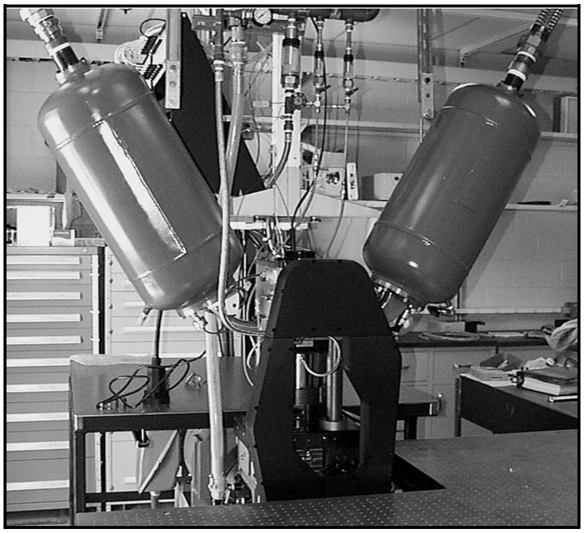

Figure 1. The HSDI Optical Engine and Laboratory

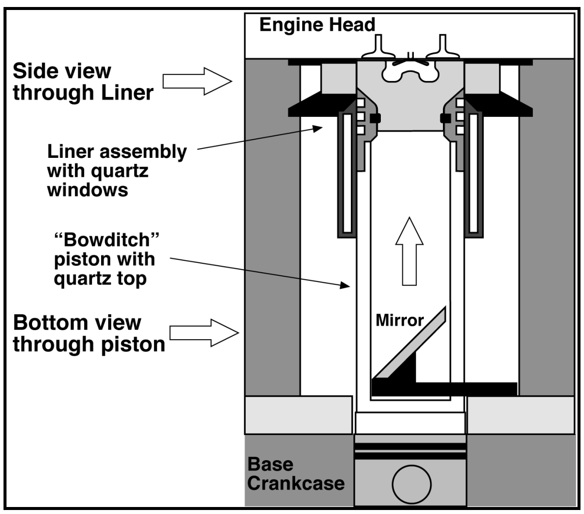

Figure 2. Schematic Demonstrating the Degree of Optical Access to the Combustion Chamber

to be influenced, and are amenable to direct experimental observation. Examples include: spray vaporization rates and injected fuel jet liquid lengths, local fuel/air ratio prior to ignition and subsequent locations of ignition and initial soot formation, and quenching of fuel or soot oxidation by walls, by convection into cold engine crevices, or by inadequate turbulent mixing.

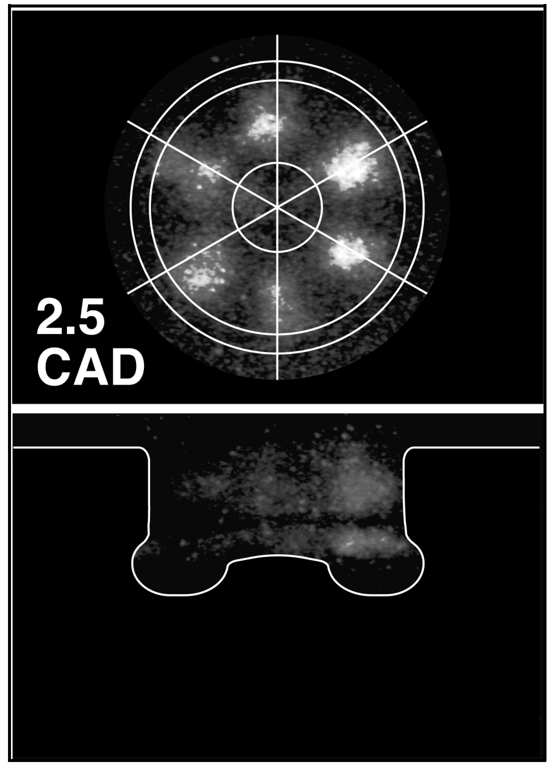

Figure 3. Early chemiluminescence observed at idle conditions. This natural radiation, associated primarily with CH and CH_2O emissions, is indicative of the onset of chemical reaction. The upper image is obtained through the piston, while the lower image is obtained through a window in the cylinder wall.

An optically-accessible HSDI diesel engine laboratory has been developed, and is shown in Figure 1. The engine is typical of state-of-the-art HSDI diesel engines which are currently being introduced into the European market-technical specifications and major features are summarized in Table 1.

The engine provides clear optical access to the combustion bowl both through a Bowditch piston arrangement, and through quartz windows in the upper cylinder liner, as seen in Figure 2. Optical access simultaneously from two orthogonal directions permits spatial ambiguities associated with line-of sight imaging to be resolved-both the axial and

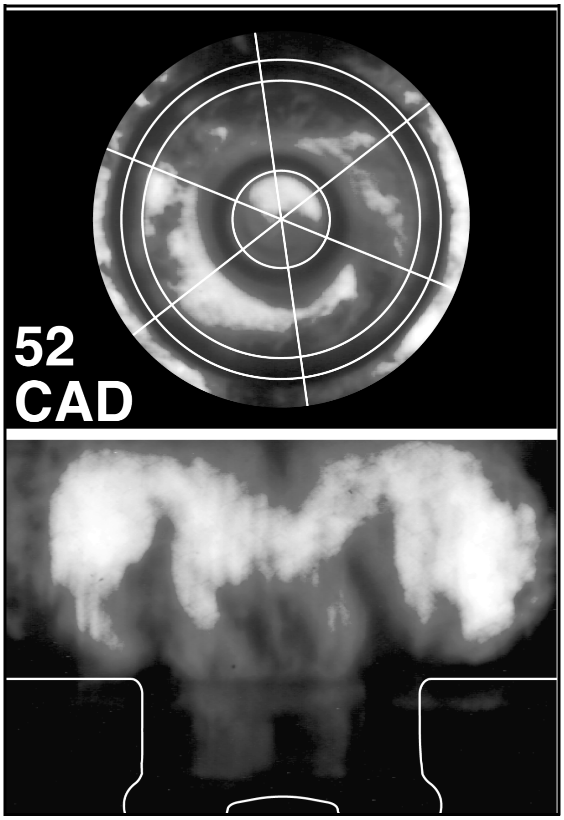

Figure 4. Natural soot luminosity observed at 2000 rpm and moderate load (5.0 bar IMEP) late in the expansion stroke. This luminosity indicates the location of hot, luminous soot which is being oxidized.

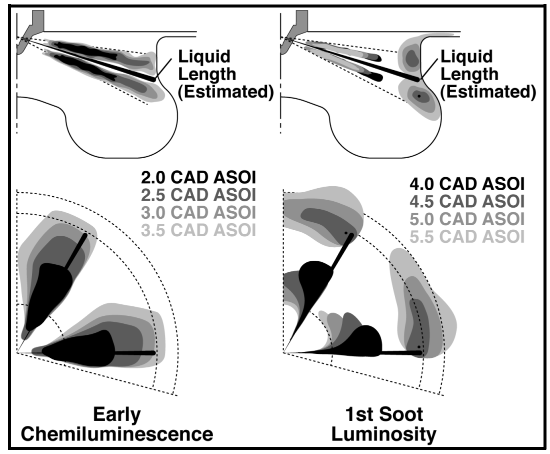

Figure 5. Schematic diagram demonstrating the evolution of the locations of early chemiluminescence and soot luminosity for the 2000 rpm, moderate load operating condition. Note the early penetration of radiant soot into the relatively cold squish volume.

radial locations of the source of radiant emissions can be determined. This enhanced spatial information is required for accurately determining ignition locations (Figure 3), initial soot spatial distributions, and regions of active oxidation during soot burnout (Figure 4). From images such as those shown in Figure 3 and 4, a physical picture of the combustion process can be generated, as is shown in Figure 5.

To effectively correlate information on the in-cylinder processes from the optical engine to measurements obtained on the traditional test engine, it is important to establish that the operating conditions of the two engines are as nearly identical as is possible. In the optical engine, slight changes geometry associated with the larger top ring land and compressive strain of the Bowditch piston result in a loss of compression. This loss is exacerbated by the increased heat transfer associated with the relatively cool optical engine combustion chamber surfaces, resulting in a reduction in cylinder pressure and bulk gas temperature. Computer modeling has been employed to select the appropriate inlet conditions such that TDC temperatures and pressures are equivalent between the two engines, as illustrated in Figures 6 and 7.

Conclusions

The optical engine has been confirmed capable of operation under typical HSDI diesel engine conditions, and imaging studies have clearly shown the evolution of the combustion process from ignition though late in the soot burnout process. Computer simulations have proven useful in supporting the experimental work; detailed comparison between the results of the simulations and the experiments are currently underway. The project is poised to continue with more quantitative, laser-based diagnostics

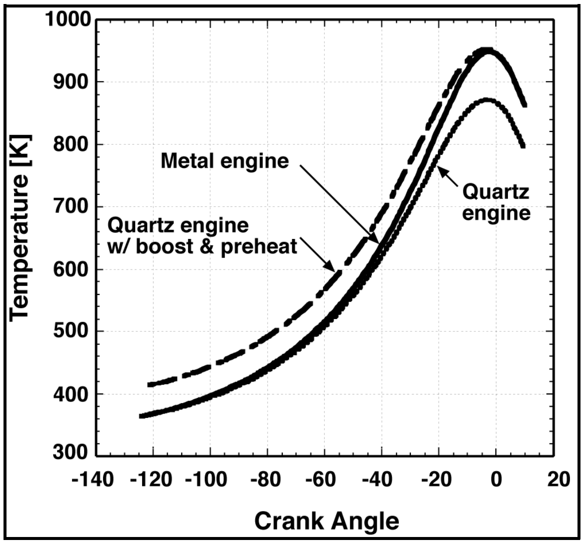

Figure 6. Computed bulk gas temperatures in the optical and traditional test engines. The upper, dashed curve demonstrates the required preheat necessary to obtain identical TDC gas temperatures.

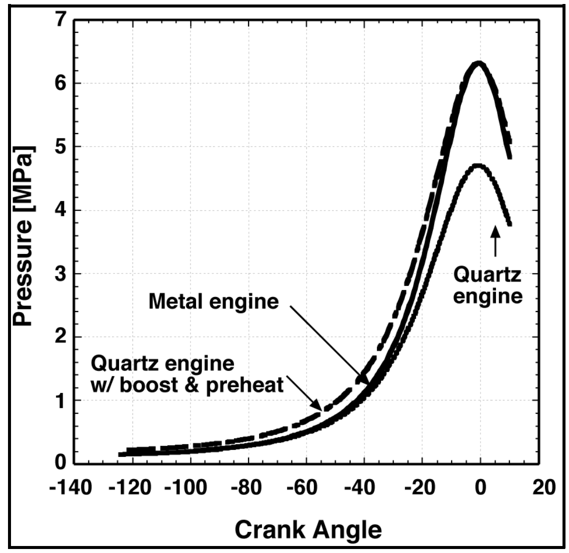

Figure 7. Computed cylinder pressure in the optical and traditional test engines. The upper, dashed curve demonstrates the additional charge pressure required to match the TDC cylinder pressures.

in the optical engine, and to complement these data with performance and emissions data obtained in the traditional test engine.

FY 1999 Publications/Presentations

Miles, P.C., "CIDI Model Development/Single-Cylinder Testing," Proc. of CIDI Research and Development Merit Review, Oct. 6-7 1998, Oak Ridge National Laboratory, Oak Ridge, TN.

Henein, N. (WSU), "Update on Wayne State University Engine Test Plan," CIDI Engine Technology CRADA Review Meeting, Nov. 19, 1998, US Council for Automotive Research, Detroit, MI.

Miles, P.C., "Progress in the Sandia Optical Engine: Injector Characterization and Chemiluminescence Imaging," CIDI Engine Technology CRADA Review Meeting, Nov. 19, 1998, US Council for Automotive Research, Detroit, MI.

Richards, K. and Reitz, R.D. (UW ERC), "Multi-dimensional Modeling of HSDI Diesel Engines," CIDI Engine Technology CRADA Review Meeting, Nov. 19, 1998, US Council for Automotive Research, Detroit, MI.

Beale, J., Kong, S., and Reitz, R.D. (UW-ERC), "Modeling Fuel Injection Using the KH-RT Hybrid Atomization Model in KIVA-3V," Joint Light-Duty/Heavy-Duty Diesel Engine CRADA Review Meeting, Feb. 2-3, 1999, Sandia National Laboratories, Livermore, CA.

Kong, S., and Reitz, R.D. (UW-ERC), "Using CHEMKIN and KIVA to Model Compression Ignition Combustion Chemistry," Joint Light-Duty/Heavy-Duty Diesel Engine CRADA Review Meeting, Feb. 2-3, 1999, Sandia National Laboratories, Livermore, CA.

Lai, M.-C. and Henein, N. (WSU), "CIDI Engine Fuel Sprays and Injection System Dynamics," Joint Light-Duty/Heavy-Duty Diesel Engine CRADA Review Meeting, Feb. 2-3, 1999, Sandia National Laboratories, Livermore, CA.

Miles, P.C., "Imaging of Natural Combustion Luminosity in the Sandia HSDI Diesel Engine," Joint Light-Duty/Heavy-Duty Diesel Engine CRADA Review Meeting, Feb. 2-3, 1999, Sandia National Laboratories, Livermore, CA.

Bianchi, G.M., Richards, K., and Reitz, R.D. (UW ERC), "Effects of Initial Conditions in Multi-Dimensional Simulations of HSDI Diesel Engines," SAE Paper No. 1999-01-1180, presented at the SAEInternational Congress and Exposition, March 1-4, 1999, Detroit, MI.

Miles, P.C., "Imaging of Natural Chemiluminescence in the Sandia HSDI Diesel Engine at Key Point III," Joint Light-Duty/Heavy-Duty Diesel Engine CRADA Review Meeting, May 5-6, 1999, Detroit Diesel Corporation, Detroit, MI.

Miles, P.C., "Characterization of the Fiat/Bosch Injection System and Comparison with the Ganser-Hydromag System," Joint Light-Duty/Heavy-Duty Diesel Engine CRADA Review Meeting, May 5-6, 1999, Detroit Diesel Corporation, Detroit, MI.

Richards, K. (UW ERC), "HSDI Diesel Engine Flow and Spray Modeling," Joint Light-Duty/Heavy-Duty Diesel Engine CRADA Review Meeting, May 5-6, 1999, Detroit Diesel Corporation, Detroit, MI.

Miles, P.C. "CIDI Model Development/Single-Cylinder Testing," Proc. DOE Diesel Combustion and Aftertreatment R&D Review, June 21-23, 1999, Argonne National Laboratory, Argonne, IL.

List of Acronyms

PNGV	Partnership for a New Generation Vehicle
CIDI	Compression-Ignition Direct-Injection
HSDI	High-Speed Direct-Injection
FTP	Federal Test Procedure
EGR	Exhaust Gas Recirculation
TDC	Top Dead Center
FIE	Fuel Injection Equipment
IMEP	Indicated Mean Effective Pressure

III.B. Diesel Combustion Cross-Cut Research

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Objectives

- Advance the scientific and technological understanding of diesel engine combustion to support the development of:
  - a new generation of diesel engines with higher efficiency and better performance that meet future emission standards.

- improved multidimensional, computational models for diesel combustion to be used in the optimization of advanced diesel engine designs.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 4, Barriers A,B

Approach

- Utilize advanced optical diagnostics coupled with a unique optically-accessible diesel combustion simulation facility to conduct these investigations.

Accomplishments

- Developed a comprehensive data base on the effects of engine, injector, and fuel parameters on diesel fuel penetration and vaporization to support DOE diesel combustion model development efforts.
- Established that fuel vaporization in a diesel spray is limited by turbulent mixing processes, not by fuel atomization processes. The results indicate that improvements are required in current diesel spray models.
- Developed a fundamentally based scaling law that provides an explanation of the effects of various parameters on diesel fuel vaporization. A simplified form of this scaling law can be used by an engine designer to assess engine, injector, and fuel properties on liquid-phase fuel penetration and vaporization.
- Relocated diesel combustion simulation facility to new lab and upgraded computer control, data acquisition, gas supply systems and hardware.

Future Directions

- Determine the effects of injector and in-cylinder conditions (including EGR) on the evolution of soot and the combustion processes in a diesel spray.
- Investigate wall impingement effects on the evolution of diesel combustion and emissions processes.
- Investigate injection rate modulation effects on diesel combustion and emissions.

Introduction

The diesel combustion cross-cut research project is focusing on improving our understanding of diesel combustion and emissions processes, and on providing a robust, well characterized database on those processes. The specific issues focused on “cross-cut” the needs of heavy-duty through light-duty diesel engine size classes. The technology base developed will support the development of advanced diesel engines, as well as multidimensional computational models for enhancing and optimizing the combustion and emissions performance of diesel engines. The goal is to determine the detailed structure of a reacting diesel fuel jet, how this structure is controlled by, and scales with, various engine operating parameters, and the evolution of the combustion and emissions formation processes. The approach is to utilize advanced, laser-based, optical diagnostics to provide this

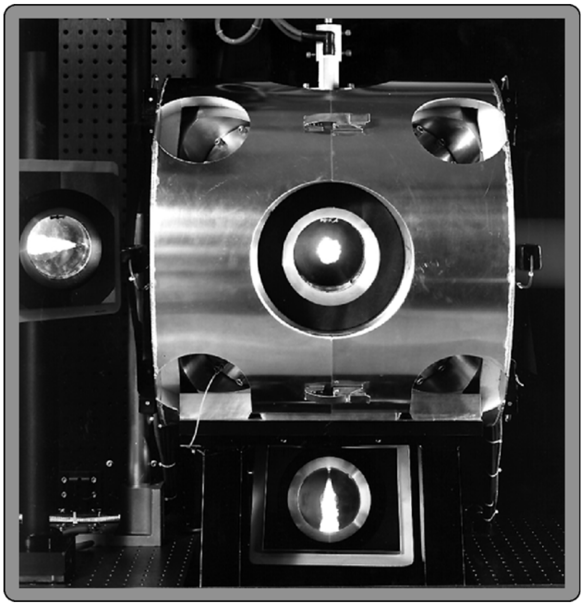

Figure 1. Photograph of the combustion vessel in operation

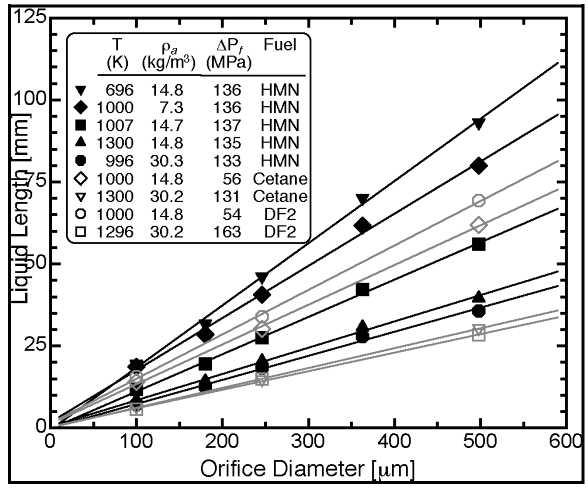

Figure 2. Liquid length versus orifice diameter for a wide range of conditions. The terms in the legend are the ambient gas temperature (T) and density (ρ_a), the orifice pressure drop (ΔP_f), and the fuel type (HMN is heptamethylnonane and DF2 is a commercial #2 diesel fuel).

information and insight.

The research is being conducted in Sandia's Diesel Combustion Simulation Facility. The diesel simulation facility consists of a constant volume combustion vessel with complete optical access. Diesel engine conditions can be simulated over a much wider range of conditions than is possible in any one engine in this facility. Conditions that can be simulated include high power density conditions with peak combustion pressures more than a factor of two higher than in present diesel engines, a direction in which most engine manufacturers would like to head for improved efficiency.

An investigation of the effects of engine, injector, and fuel parameters on diesel fuel penetration and vaporization processes in a direct-injection (DI) diesel engine was completed this year. Liquid-phase fuel penetration and evaporation are important factors in optimizing DI diesel engine combustion processes, especially for the small-bore automotive diesels presently under development. Penetration of the liquid-phase fuel is needed to promote fuel-air mixing, but can lead to greater emissions if the liquid fuel impinges and collects on piston bowl walls. As a result, understanding how various parameters affect the penetration of liquid-phase fuel and which processes control fuel vaporization in a diesel spray are important, both to the engine designer and to those developing multi-dimensional computational models for use as engine design tools.

In addition, during the past year the diesel combustion simulation facility was moved to a new laboratory and upgraded.

Results

The diesel spray fuel vaporization and penetration research was performed in the Diesel Combustion Simulation Facility (DCSF) using an electronically controlled, common-rail diesel fuel injector. The range of conditions that can be covered in this facility is unique in the world, and includes conditions in current

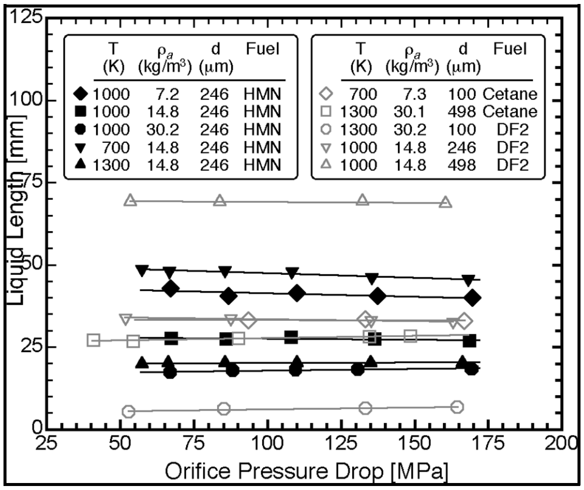

Figure 3. Liquid length versus the pressure drop across the injector orifice for a wide range of conditions. The term d in the legend is the orifice diameter.

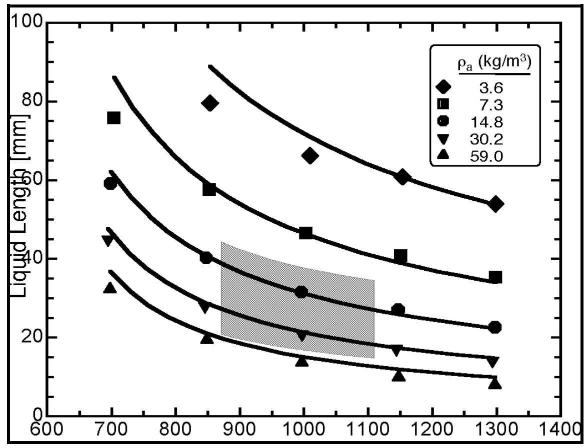

Figure 4. Cetane liquid length as a function of gas temperature for five gas densities (ρ_a). The symbols are measured data and the curves are given by the scaling law [Siebers, 1999]. The orifice pressure drop, the orifice diameter, the ambient gas temperature, and the fuel temperature were 136 MPa, 246 μm, and 438 K, respectively. The light gray region in the figure represents the range of liquid lengths expected in light- and heavy-duty DI diesels.

and proposed, advanced diesel engines. Figure 1 shows the picture of the optically accessible combustion vessel in the DCSF. Parameters varied in the investigation included: injection pressure, orifice diameter and aspect ratio, ambient gas temperature and density, and fuel temperature and volatility.

Fuel penetration and vaporization processes were investigated through measurements of the maximum penetration distance of liquid-phase fuel. This maximum penetration distance, referred to as the liquid length, is a very sensitive measure of the fuel vaporization process. Images of Mie-scattered light from the liquid-phase fuel in diesel sprays were used to determine the maximum penetration distance of the liquid-phase fuel.

Two of the most revealing trends observed in spray. This conclusion was arrived at by considering the expected dependence of liquid length on orifice diameter and injection pressure in two limiting cases of fuel vaporization: (a) control by air entrainment and mixing and (b) control by local interphase transport processes (e.g., heat and mass transfer at droplet surfaces). Only the former explains the trends in Figures 2 and 3.

Based on the concept of mixing-limited vaporization, a scaling law was developed for liquid-phase fuel penetration in diesel sprays [Siebers,1999]. The scaling law accounts for the effects of injector, fuel, and in-cylinder conditions on liquid length. Comparison of the scaling law to measured data shows that scaling law reproduces the trends observed in the experimental data with respect to all parameters considered. Figures 4 and 5 present comparisons of the scaling law with measured data as a function of ambient gas temperature and density for two single-component fuels, cetane and heptamethylnonane. The two fuels span a significant fraction of the volatility range of standard diesel fuel.

An implication of mixing-limited vaporization is that the processes of atomization and the ensuing interphase transport of mass and

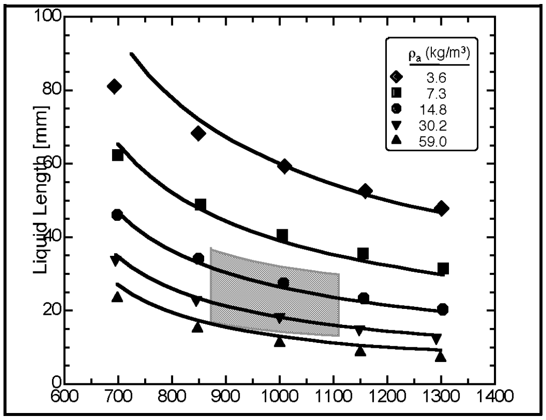

Figure 5. Heptamethylnonane liquid length as a function of gas temperature for five gas densities.

energy at droplet surfaces are not the controlling processes with respect to fuel vaporization in DI diesel sprays. This further implies that better atomization (i.e., smaller droplets) alone will not promote increased fuel vaporization.

The results also suggest that current diesel spray models need to be improved. The basic formulation of current models does not allow the gas-phase flow (i.e., the air entrainment and turbulent mixing) in the fuel vaporization region of the spray to be adequately resolved. This deficiency makes modeling of the combustion and emissions formation processes further downstream in the spray difficult.

A major focus this year was also on relocating the DCSF to a laboratory in the new section of the Combustion Research Facility. As part of this move the facility was upgraded. All the facility systems (e.g., the data acquisition and control computers and instrumentation, the fuel injection system, the gas supply system, the lasers, and the combustion vessel) are now operational. The remaining task before the relocated facility is fully operational is the installation of a safety shield around the combustion vessel to protect personnel against a window failure. The new location and upgrades provide for more efficient use of technician support, significantly improved laboratory facilities, improved data acquisition and control computers, and faster data throughput.

Conclusions

A comprehensive understanding of the effects of engine, injector, and fuel parameters on diesel fuel penetration and vaporization processes was developed, along with a supporting data base and fuel penetration scaling law. The data base and scaling law provides a fundamental baseline on liquid fuel penetration and vaporization in diesel sprays that can be compared with the vaporization aspects of the multi-dimensional diesel spray models under development. The scaling law can also provide design guidance on the expected maximum extent of liquid-phase fuel penetration in diesel engines.

The results show that fuel vaporization in a diesel spray is controlled by turbulent mixing processes in a diesel spray. Atomization and transport processes at the surface of droplets do not limit fuel vaporization, as was previously thought. The results further indicate that improvements are required in current diesel spray model formulations to account for air entrainment and turbulent mixing processes in the near injector region of the spray.

In addition, the diesel combustion simulation facility was relocated and is nearly operational. The remaining task is the installation of a safety shield around the combustion vessel to protect personnel against a window failure.

Over the remainder of FY99, an investigation of the effects of injector and in-cylinder conditions (including EGR) on the evolution of soot in a diesel spray will be initiated. The goal will be to establish the mechanisms by which parameters such as orifice diameter, fuel injection pressure, and in-cylinder conditions affect soot production in a diesel engine.

References/Publications

D. L. Siebers, "Scaling Liquid-Phase Fuel Penetration in Diesel Sprays Based on Mixing-Limited Vaporization," Paper No. 1999-01-0528, SAE International Congress, Detroit, MI, February, 1999.

B. Higgens, C. Mueller, and D. Siebers, "Measurements of Fuel Effects on Liquid-Phase Penetration in DI Sprays," Paper No. 1999-01-0519, SAE International Congress, Detroit, MI, February, 1999.

D. L. Siebers, "Liquid-Phase Fuel Penetration in Diesel Sprays," Accepted to the Transactions of the SAE (Paper No. 980809, SAE International Congress, Detroit, MI, February, 1998).

List of Acronyms

DCSF	Diesel Combustion Simulation Facility
DI	Direct injection
EGR	Exhaust Gas Recirculation

III.C. Near-Field Characterization of Diesel Sprays Using X-Rays

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Objective

- Gain a thorough understanding of spray breakup and atomization processes in the near-field region of diesel sprays. An understanding of these phenomena could lead to better spray atomization, reduced engine emissions, and improved spray nozzle geometry.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 6, Barriers A,B,C

Approach

- Establish a thorough understanding of the nature of the spray core:
  - air entrainment, spatial distribution of mass flux, and local A/F ratios;
  - spray axial parameters (cone angle, tip penetration); and
  - drop sizes and number densities.
- Study the mechanisms leading to spray breakup and atomization.
- Investigate the effects of nozzle geometry and injection parameters on near-nozzle and far-downstream spray characteristics; this information is of interest to nozzle designers.
- Use facilities at the Advanced Photon Source (APS), the world's brightest X-ray source, for this study. Many features of this facility—monochromaticity, tunable X-ray source, high collimation, and unique detection systems—are ideally suited for achieving the stated objective.

- Use techniques of X-ray absorption, small-angle X-ray scattering, and fluorescence to measure important spray characteristics in the near-field region, such as liquid core length, air/fuel ratio, air entrainment, drop size, and velocity.

Accomplishments

- An initial set of experiments for X-ray absorption in a diesel spray was completed. The diesel spray for these measurements was determined by using a Bosch common rail system while injecting the fuel into a high-density gas environment.
- By performing point measurements, such characteristics as liquid core length, local air/fuel ratios, and spray tip velocities in the near-field region were established.

Future Directions

- Improve spatial and temporal resolution of X-ray absorption measurements by using advanced detection concepts.
- Perform further measurements by using Small Angle X-ray Scattering and Fluorescence.
- Postulate a theoretical model for diesel spray formation on the basis of observations.

Introduction

Many regions of a typical diesel spray, especially those in the near-field region, have remained impenetrable to conventional laser diagnostics (cf. Regions I & II in Figure 1). Processes occurring in these regions have a significant bearing on downstream atomization and overall emissions performance. The present effort focuses on overcoming the limitations of conventional laser diagnostics by using highly penetrating X-rays to obtain a comprehensive picture of spray formation.

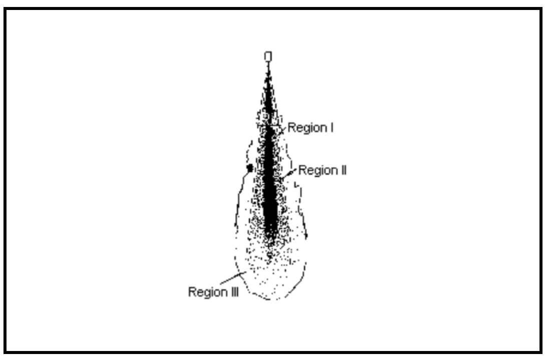

Figure 1. In a typical diesel spray, central regions I and II are impenetrable to conventional laser diagnostics

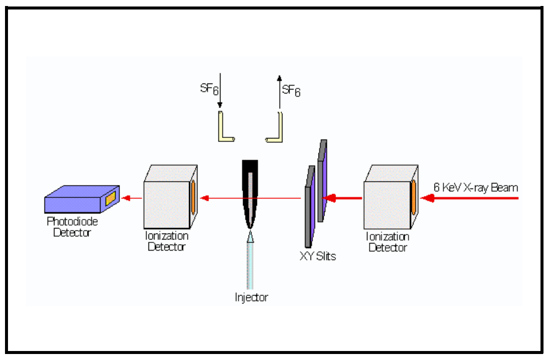

Figure 2. A schematic of the experimental setup to perform X-ray absorption

In addition to their highly penetrating nature, a number of features unique to X-rays prove attractive for spray characterization. These are summarized in Table 1.

The proposed effort consists of two tasks:

> **Task I**: Use conventional diagnostics to establish baseline spray characteristics to help conduct experiments in task II.
> **Task II**: Use X-ray diagnostics to probe optically impenetrable regions of diesel

Table 1. Potential Capabilities of X-Rays for Spray Characterization

Technique	Potential Capabilities	Advantages over Laser Diagnostics
Absorption	Liquid core length, local (F/A)	• High penetrating capacity to yield measurements in optically opaque regions
Small Angle X-ray Scattering (SAXS)	Drop size and velocity	• Measurement capability in optically opaque regions
X-ray fluorescence	Tracing of fuel mass, even in combusting sprays	• Atomic fluorescence as opposed to radical/molecular fluorescence; fluorescence spectra are element-specific • Can yield quantitative measurements

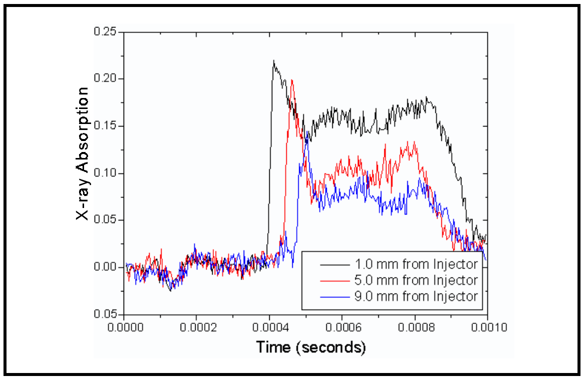

Figure 3. Measured absorption profiles 1 mm, 5 mm, and 9 mm downstream of the injector

sprays. Unique facilities at the Advanced Photon Source (APS), the world's brightest X-ray source, will be used.

Task I Results: Correlations were obtained for spray tip penetration, S (mm), and spray cone angle, Θ, through high-speed imaging of diesel sprays in a quiescent high-pressure chamber.

$$S \propto t^{0.733} \triangle P^{0.257} \rho_a^{-0.29}$$

$$\tan(\Theta/2) \propto t^{-0.39} \triangle P^{-0.13} \rho_a^{0.19}$$

Such correlations were used for designing and sizing of components for experiments in task II.

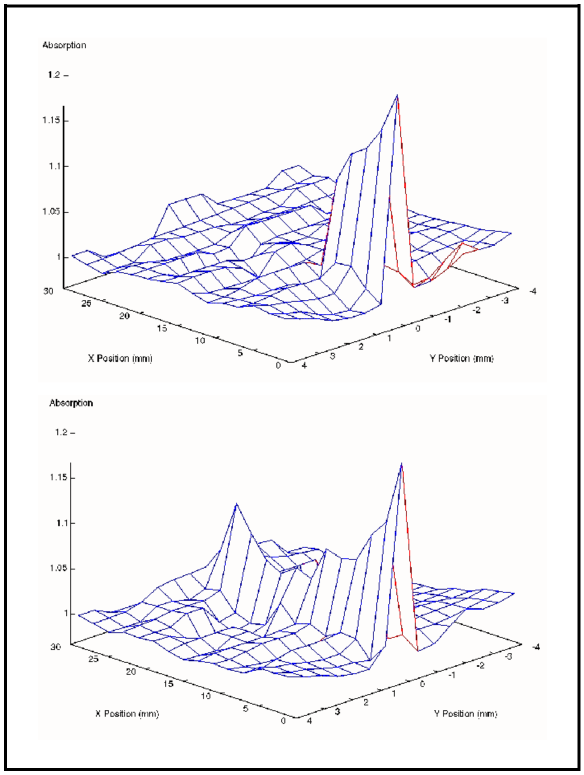

Figure 4. Reconstructed absorbance 71 µs and 176 µs after the start of injection

Task II Results: X-ray absorption experiments were conducted in an experimental setup, as shown in Figure 2. Such measurements enable estimation of fuel mass in the path of the beam, liquid core length, spray tip velocity, and local air/fuel ratio. Preliminary results from such measurements are presented below.

Measured absorption profiles 1 mm, 5 mm, and 9 mm downstream of the injector are shown in Figure 3. An initial peak followed by two bumps (as shown in this figure) was observed in all measurements. Interpretation of such observations in terms of physical processes is under way. Also, a constant value for spray tip velocity (~127 m/s) obtained through such measurements confirms the use of a low ambient density of ~ 6 kg/m^3.

Reconstructed absorbance profiles at two instants after the start of injection are shown in Figure 4. The time evolution of local air/fuel ratio, liquid core length, and air entrainment in the near-field region could be captured through a sequence of such profiles. Further insights are likely through future measurements of SAXS.

While the rest of the data are being processed, it ought to be noted that this is a successful demonstration of the use of X-rays for diesel spray characterization. More measurements encompassing variation of injection parameters are scheduled.

Publications/Presentations

S. Gupta, R. Poola, R. Sekar, J. Wang, C. Powell, "Near-Field Characterization of Diesel Sprays Using x-rays," presented at the annual DOE Diesel Combustion and Aftertreatment R&D review, June 21-23, 1999, Argonne, IL.

"X-ray Diagnostics for Diesel Sprays," Submitted to ASME - ICE spring technical conference.

List of Acronyms

APS Advanced Photon Source
SAXS Small Angle X-ray Scattering

III.D. EGR Optimization : Integrated Plan

Overall Objective

The following three subtasks all focus on developing a fundamental understanding of the processes which define the practical limit for how much emissions reduction can be achieved through exhaust gas recirculation (EGR). Such information is crucial for determining the limits of available EGR technology and what new EGR-related technologies must be developed for CIDI engines to meet current and anticipated emissions requirements. Ultimately, it is desirable to maximize the emissions reducing potential available from EGR.

Task Integration

As shown in the flowchart in Figure 1, three individual subtasks have been planned so that they combine to form an efficient strategy for addressing the expected dominant physical processes linking EGR and emissions. The two major challenges to EGR optimization are 1) non-uniform mixing and distribution of the EGR charge and 2) the impact of these EGR non-uniformities and variations in other parameters on combustion stability.

Mixing and distribution non-uniformities are associated with the complex turbulent flow in the EGR and intake air systems. Subtasks 1 and 3 are designed to provide fundamental quantitative information regarding the degree of non-uniformity and how this correlates with engine operating

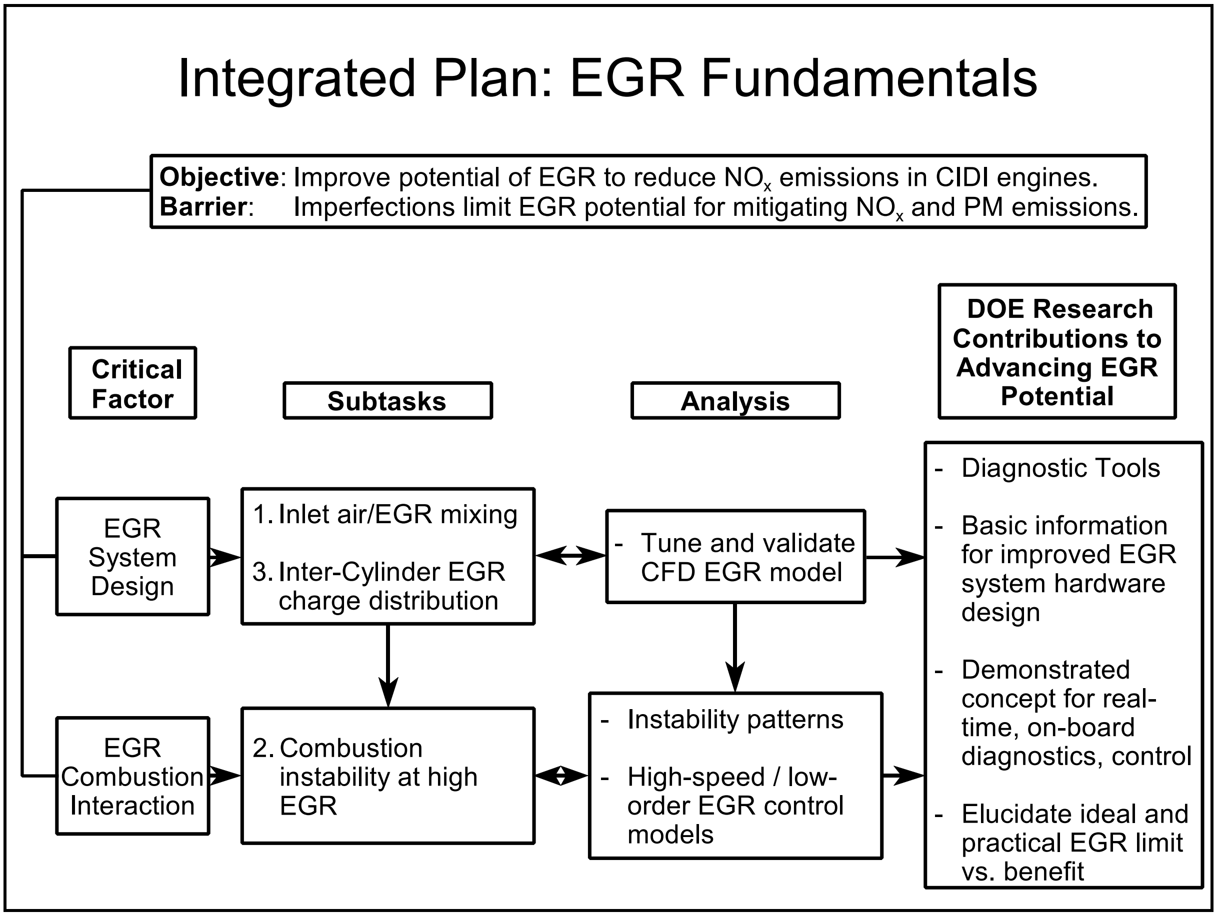

Figure 1. Flowchart Showing Relationship Among Subtasks 1, 2, and 3

conditions and EGR parameters. The approach involves direct experimental measurements of the non-uniformities in a commercial CIDI diesel engine using advanced optical techniques. These measurements will be used to develop a state-of-the-art commercial computational-fluid-dynamics (CFD) model of the EGR system, which will allow industry to assess their internal codes and realize improved EGR-system design. Moreover, results from these two subtasks provide important inputs to subtask 2.

Combustion instability is ultimately the reason that an upper limit exists for EGR. Specifically, the in-cylinder combustibility of the fuel is reduced by the addition of successively higher levels of EGR, up to the point that combustion becomes so poor that particulate levels are excessive and energy efficiency deteriorates. Variations among cylinders and between engine cycles cause the practical EGR limit to be significantly lower than would be possible under ideal circumstances. Subtask 2 is designed to address the connection between EGR non-uniformities, combustion quality, emissions, and engine operation. This subtask utilizes special tools from nonlinear dynamics theory, with experimental measurements of cycle-resolved combustion and emissions from the same commercial engine used in subtasks 1 and 3, to gain insight into combustion instability. The results of subtask 2 will provide the resources to extend the EGR potential via implementation of advanced control strategies incorporating phenomena related to combustion stability.

As shown in Figure 1, it is expected that the results of these three tasks will lead to both improvements in EGR design and control strategies and a clearer delineation of the practical potential of EGR for NO_x reduction.

III.D.1. EGR Optimization Subtask 1: Measurements of EGR Distribution and Mixedness in a CIDI-Engine Intake

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Objectives

Develop diagnostic capability to characterize the EGR/air distribution and mixedness in the intake manifold of an operating CIDI engine. Apply diagnostics to develop a database for tuning and validation of an industry-standard CFD model of the EGR process. The validated model is to be used by industry to improve the potential of EGR to reduce NO_X emissions in CIDI engines by affording improved EGR system designs.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 3, Barriers A,B

Approach

Use planar laser-induced fluorescence (PLIF) of molecularly-seeded exhaust to quantitatively resolve the degree of mixing between EGR and air.

Accomplishments

- Bench measurements indicate excellent accuracy of the diagnostic over the range of environmental conditions existing in the intake manifold.
- Established industrial/agency/laboratory consensus on standardized test cases for model validation and to demonstrate model adaptability.
- Developed integrated EGR plan.
- Established industrial consensus on CFD model representing an industry standard.
- Fabricated and tested optically-accessible intake manifold
- Acquired equipment for high-speed PLIF imaging.

Future Directions

- Complete bench measurements.

- Demonstrate high-speed PLIF system.
- Implement diagnostic in intake manifold of running CIDI diesel engine.
- Apply diagnostic for measurements at test cases and higher-order cases as necessary.

Bench measurements have been used to verify the accuracy of the PLIF measurements using the instrumentation shown in Figure 1. These measurements have demonstrated the linearity of the signal with molecular seed concentration. No interferences have been identified using major constituents of exhaust and n-octane and propylene to represent unburnt hydrocarbons. Furthermore, the bench measurements have indicated that electronic quenching does not influence the measurements over the range of environmental conditions existing in the intake manifold. This result is supported by data indicating dominance of the molecular-seed's dynamics by inter-system crossing. This disparity of rates which effectively mitigates electronic-quenching issues should allow the accuracy of the PLIF diagnostic to rival that of more fundamental diagnostics such as absorption. Thus, the PLIF images should afford an accurate measure of local EGR concentration and degree of EGR/air mixedness.

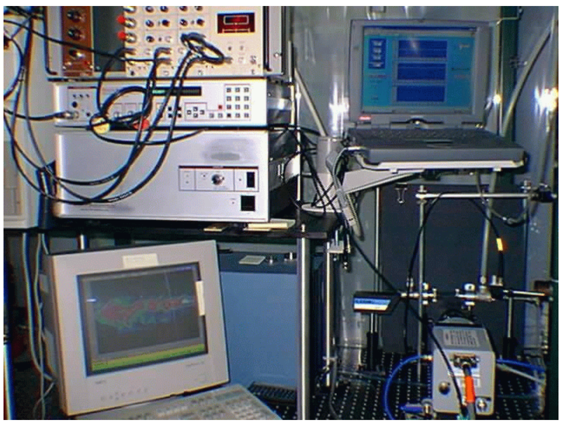

Figure 1. Bench Setup for the PLIF and Point Measurements

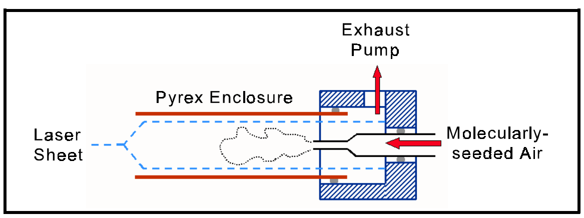

Figure 2. Schematic of Test Cell for Multiple Gas Mixing

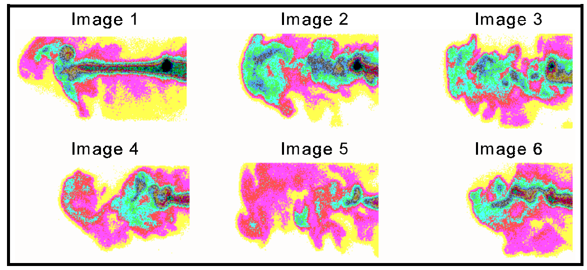

Figure 3. Single-shot PLIF Images of Tagged Air Jet Injection into Counter Flowing Fresh Air Stream at Steady State

Using bench measurements and a test cell of dimensions representative of the relevant intake manifold, PLIF images of multiple gas-stream mixing have been made. These were single-shot measurements using a gate width of 20 ns to effectively freeze the flow. As indicated in Figure 2, the test cell consists of a molecularly-seeded air jet propagating from right to left, representing the EGR. This tagged jet is injected into a counter flowing stream of fresh air flowing from left to right in the figure. The two gas streams mix in a space confined by a Pyrex tube and are exhausted out the right-hand side of the test cell by a pump. A vertical laser sheet is injected to bisect the tagged jet, and PLIF images are acquired normal to this sheet.

Six single shot images of the multiple-gas mixing in the test cell at a steady-state operating condition are show in Figure 3.

These images demonstrate the ability to assess local EGR concentration, degree of mixedness, flow structures, and shot-to-shot fluctuations in the mixing. Similar fluctuations in the mixing are likely to exist in the CIDI diesel engine and could degrade the system stability and ultimate EGR potential. We are working to identify an optimum scalar parameter to characterize the mixedness in a given single-shot image. This parameter will be useful, along with synchronous measurements of other system parameters, to quantify the coupling and propagation of mixedness fluctuations through the system.

List of Acronyms

CIDI Compression Ignition Direct Injection
CFD Computational Fluid Dynamics
EGR Exhaust Gas Recirculation
PLIF Planar Laser-Induced Fluorescence

III.D.2. EGR Optimization Subtask 2: Extending Exhaust Gas Recirculation Limits in Multicylinder CIDI Engines

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CRADA Partner: Ford Motor Company, Dearborn, Michigan
CRADA No. ORNL 95-0337
Ford Investigators: John Hoard, Lee Feldkamp, Tony Davis, Frank Connolly
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Objective

- Develop options for extending practical EGR limit to further reduce NO_X.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 3, Barriers A,B

Approach

- Identify correlation between EGR operating and system parameters and combustion instability and emissions.
- Develop fundamental understanding of non-random patterns in combustion instability at high EGR.

- Exploit understanding of non-random combustion patterns and effects of non-uniform EGR mixing and distribution (see parts 1 and 3) to develop options for improving EGR design and control.

Accomplishments

- Two sets of preliminary high EGR tests completed on the 1.2-L, 4-cylinder Ford DIATA (Direct Injection, Aluminum, Through-bolt Assembly) diesel engine.
- Above Ford tests confirm suspected onset of significant non-random patterns in combustion stability associated with high EGR.
- Correlations between combustion variations and emissions indicate the possibility of predicting emissions from combustion parameters which in turn can potentially be derived using existing engine sensor technology.
- Similar observations in spark-ignition engines have led to demonstration of the first-ever cycle-by-cycle control that reduces combustion variability without changing the mean engine calibration.
- Low-order model for lean-combustion cyclic variability in spark-ignition engines successfully modified to account for the general observed trends with EGR.
- 1.9-L, 4-cylinder VW engine at ORNL configured to operate with adjustable EGR.
- ORNL VW engine outfitted with LBNL scatterometer, Cambustion fast-FID analyzer, Kistler/Optrand in-cylinder pressure transducers, and Real-Time Engineering data acquisition system.
- Combustion fast-NO_X analyzer ordered for VW engine testing.

Future Directions

- Cycle-resolved measurements of in-cylinder pressure, particulate matter, NO_X, and hydrocarbons on ORNL VW engine for range of EGR levels.
- Develop low-order dynamic model linking high EGR diesel combustion instability with emissions.
- Develop concepts for on-board diagnostics capable of monitoring diesel combustion instability and inferring emissions with existing (or minimally improved) engine sensors.
- Relate combustion instability to non-uniformities in EGR mixing/distribution
- Develop EGR design/control strategies for meeting NO_X, particulate goals.

Introduction

This activity builds on an earlier collaboration between ORNL and Ford under a pre-existing CRADA (ORNL 95-0337). Under the original CRADA, the principal objective was to understand the fundamental causes of combustion instability in spark-ignition engines operating under lean conditions. The results of this earlier activity demonstrated that such combustion instabilities are dominated by the effects of residual gas remaining in each cylinder from one cycle to the next. A very simple, low-order model was developed that explained the observed combustion instability as a noisy nonlinear dynamic process. The model concept lead to development of a real-time control strategy that could be employed to significantly reduce cyclic variations in real engines using existing sensors and engine control systems. Figure 1 illustrates the effectiveness of this control in a commercial engine. The combustion indicator has been normalized to preserve

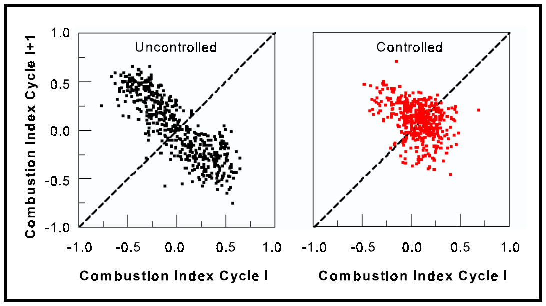

Figure 1. Experimental Example of Reduction in Combustion Instability Achieved on a Commercial Spark-Ignition Engine with Cycle-Resolved Feedback Control

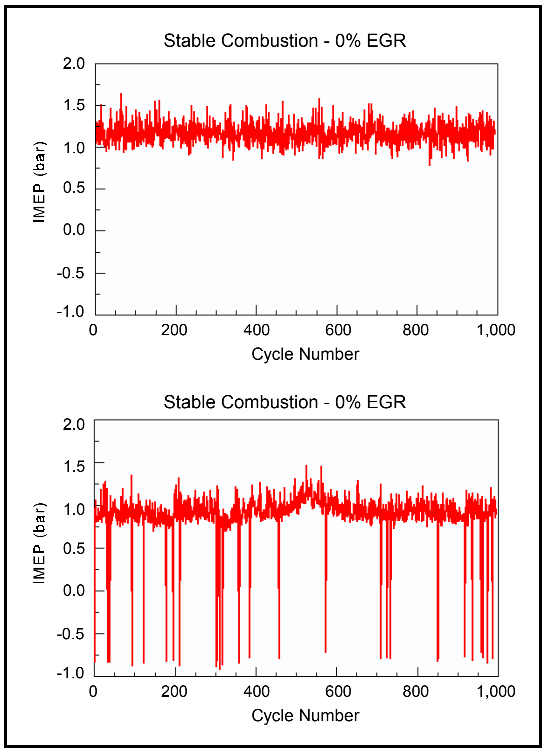

Figure 2. Examples of "Normal" and High EGR Combustion Variability Observed with DIATA Engine

CRADA-protected information.

With funding from OAAT, the CRADA has been modified to focus more on EGR and CIDI engines. The modified CRADA now includes engine experiments and analysis at both Ford and ORNL.

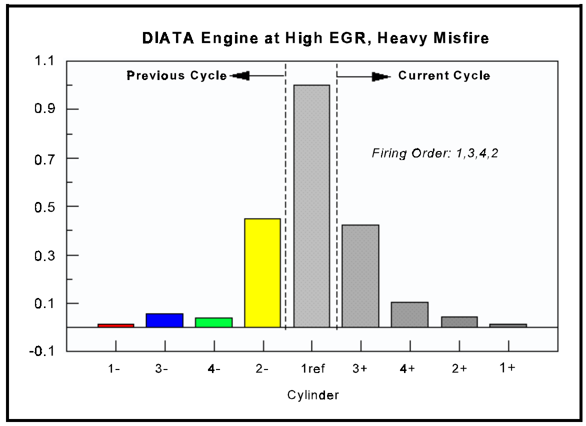

Figure 3. Example of Correlation Between Combustion Indifferent Cylinders in the DIATA at High EGR

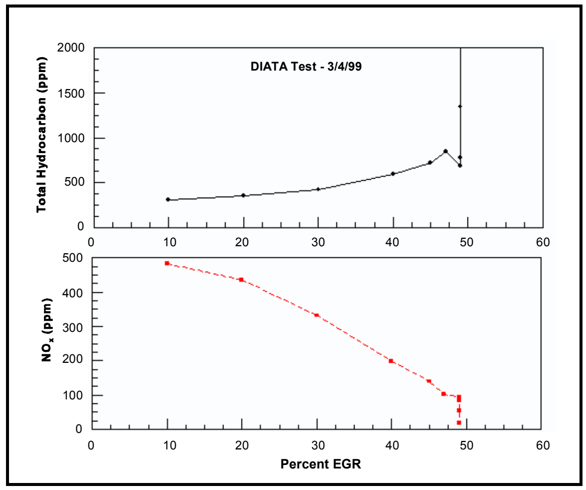

Figure 4. Example Correlation Between Hydrocarbon and NO_X Emissions for the DIATA Engine with Increasing EGR

Activities at Ford

Example combustion instability observations from the preliminary Ford DIATA experiments are illustrated in Figure 2. Under very high EGR conditions we observed that combustion became so unstable that complete misfires occurred frequently. Variations in the performance of each cylinder also indicated significant non-uniformity in EGR distribution.

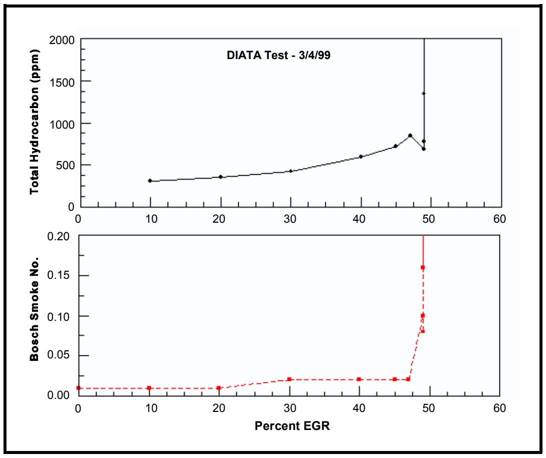

Figure 5. Example Correlation Between Hydrocarbon Emissions and Smoke for the DIATA Engine with Increasing EGR

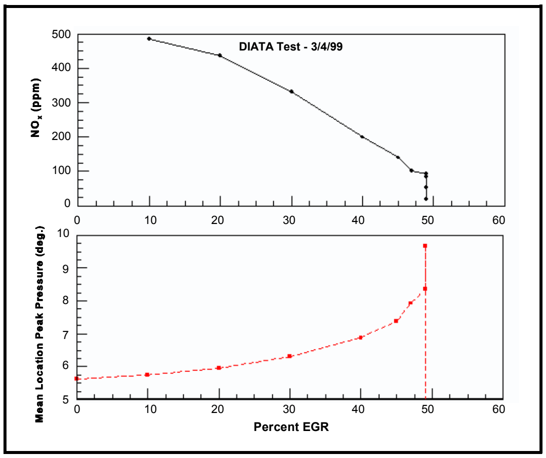

Figure 6. Example Correlation Between Combustion and Emissions for the DIATA Engine with Increasing EGR

The non-random aspects of the observedcombustion instability are illustrated in Figure 3. Here we observe that events in cylinders firing successively in order were significantly correlated. This suggests cylinder-to-cylinder communication (possibly via EGR non-uniformities) and potential for feedforward control.

The strong correlations between different emission species and combustion and emissions seen with the DIATA are illustrated in Figures 4-6. We observe that NO_X, hydrocarbons, and smoke track consistently with EGR, and

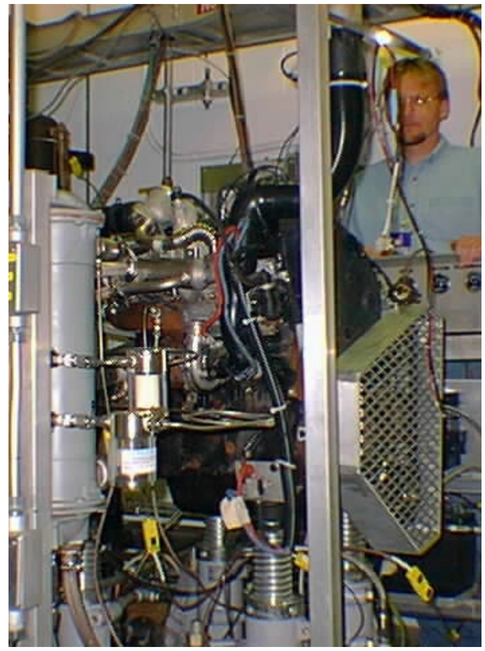

Figure 7. Volkswagen 1.9-L, Turbo-charged, Direct-injection Engine Used for ORNL EGR Experiments

these in turn correlate with the combustion timing (as reflected in the location of the peak cylinder pressure or LPP). Note that at high EGR at point is reached where very small EGR changes have a highly nonlinear effect.

Activities at ORNL

ORNL experiments with the 1.9-L VW engine (Figure 7) are expected to begin in July of 1999. The focus of these tests will be to confirm and extend the observations with the Ford DIATA. With the advanced diagnostics equipment now being installed, it will be possible to develop a much more direct connection between combustion variations and emissions.

Analysis of previous of previous spark-ignition data with EGR has led to a modification of the original low-order combustion instability model to account for EGR effects. This model is now being used as a basis for developing a low-order CIDI model.

Summary

Preliminary engine experiments have demonstrated that nonrandom combustion instability patterns occur in diesel engines as well as spark-ignition engines at high EGR. The analytical tools developed for spark-ignition engines appear to be useful for diagnostics of combustion instabilities in diesels. A strong correlation appears to exist between certain pressure-derived combustion parameters and emissions. This has significant implications for using existing sensors to infer engine emissions.

Highly detailed measurements of cycle-resolved combustion and emissions are planned for ORNL VW engine in the very near future.

References/Publications

Green J.B. Jr., Daw C.S., Armfield J.S., Finney C.E.A., Wagner R.M., Drallmeier J.A., Kennel M.B., Durbetaki P. (1999). "Time irreversibility and comparison of cyclic-variability models", SAE Paper No. 1999-01-0221.

Daw C.S., Kennel M.B., Finney C.E.A., Connolly F.T. (1998). "Observing and modeling nonlinear dynamics in an internal combustion engine", Physical Review E 57:3, 2811-2819.

Finney C.E.A., Green J.B. Jr., Daw C.S. (1998). "Symbolic time-series analysis of engine combustion measurements", SAE Paper No. 980624.

Wagner R.M., Drallmeier J.A., Daw C.S. (1998). "Prior-cycle effects in lean spark ignition combustion: fuel/air charge considerations", SAE Paper No. 981047.

Wagner R.M., Drallmeier J.A., Daw C.S. (1998). "Nonlinear cycle dynamics in lean spark ignition combustion", 27th International Symposium on Combustion (Boulder, Colorado USA; 1998 August 2-7).

List of Acronyms

CIDI	Compression Ignition Direct Injection
CRADA	Cooperative Research and Development Agreement
DIATA	Direct Injection, Aluminum, Through-bolt Assembly
EGR	Exhaust Gas Recirculation
LBNL	Lawrence Berkeley National Laboratory
LPP	Location of Peak Cylinder Pressure
OAAT	Office of Advanced Automotive Technologies
ORNL	Oak Ridge National Laboratory
VW	Volkswagen

II.D.3. EGR Optimization Subtask 3: Measurements of the Intake Manifold Mixing and Transport of Recirculated Exhaust Gas in a High-Speed, DI Diesel Engine

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Objectives

Using measurements of the cylinder-to-cylinder EGR distribution during both steady operation and engine transients, create a database to be used in the comprehensive validation of a numerical, CFD engine simulation model of a high-speed, DI Diesel engine.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 3, Barriers A,B

Approach

- Conduct experiments in a 'generic' production engine using laser-absorption spectroscopy to measure the local concentration of recirculated exhaust gas flowing into the intake ports.
- Generate an experimental database of cycle-resolved, ensemble-averaged measurements during both steady operation and engine transients.
- Use an engine-simulation code coupled to a 3-D CFD code to correlate the experimental measurements with the engine operating conditions.
- Validate and verify the simulation capability of the codes in predicting the cylinder-to-cylinder distribution of EGR.

Accomplishments

- Volkswagen 1.9L TDI engine is set up and is fully operational in the laboratory, with the capability of computer control under either steady conditions or speed/load/EGR transients.
- The optical access to the intake ports is fabricated, installed and operational.
- The infrared, diode-laser based CO_2 absorption diagnostic is fully setup and operating in the engine. Data acquisition can be performed during both steady operation and transients, yielding the required cycle-resolved, ensemble-averaged data.
- Two suites of engine-simulation/CFD computer codes are being considered for the correlation of the experimental database.

Future Directions

- Comprehensive data acquisition and the database construction will begin in mid-July and the experimental database will be continuously updated during the experimental work.
- Engine simulation and CFD codes will be used for an iterative verification of the modeling adequacy.
- Compare the measurements made using the optical diagnostic with similar measurements using a sampling technique—currently used by industry during engine development.
- Validated simulation model of EGR mixing and distribution is available for EGR- system design and development.
- Perform an assessment of the validated simulation model to correctly predict the performance of hardware that has been purposely modified to create a mal-distribution of EGR.

Introduction

The new generation of small-bore, high-speed, direct-injection diesel engines being developed for automotive applications are expected to use large amounts of EGR to control the emission of NO_X. This, in turn, may lead to problems involving the mixing of EGR with fresh air in the intake manifold and cylinder-to-cylinder distribution of EGR. To better our understanding of this problem and the underlying processes, we have devised an experimental research program that will result in a database of experimental measurements which will allow the validation and verification of CFD simulations of the mixing process.

Engine

The experimental work will be conducted using a Volkswagen 1.9 liter TDI diesel engine. This engine design incorporates much of the current technology that is being used in the development of domestic diesel engines for automotive applications, and is therefore a good choice as a 'generic' engine for this study.

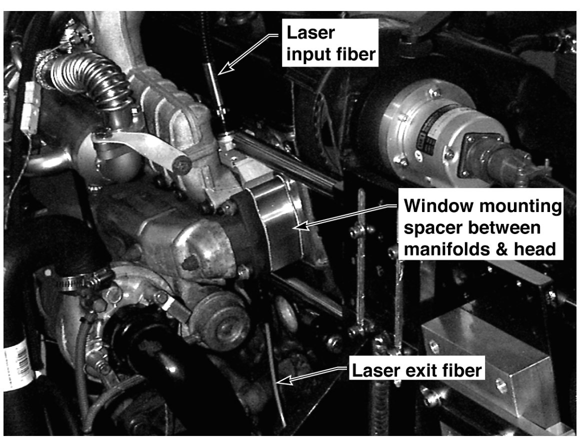

Figure 1. The optical spacer plate is located between the cylinder head and the manifolds. The fluoride-glass fibers carry the laser light from the laser to the engine, then back to the In-Sb detector.

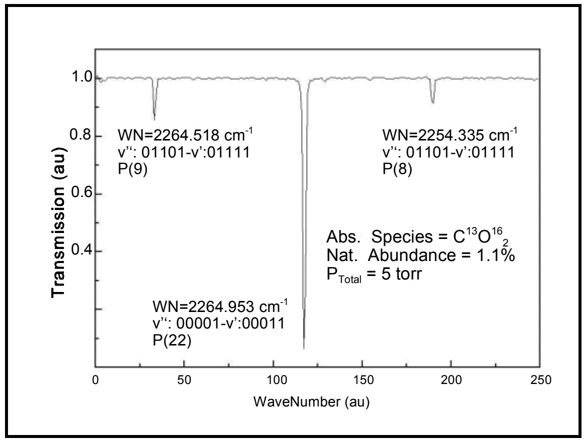

Figure 2. A spectral scan through CO_2 at 5 torr in a 10 cm. reference cell illustrating the spectral region being used in this study.

The engine has been mounted on an engine stand and fitted with a water brake dynamometer.

The engine operation is primarily controlled through the Volkswagen ECM, with the exception of the accelerator positioner and the EGR control valve. We have configured a lab computer to control engine speed, load and EGR in such a way that we can operate under steady speed, load, and EGR conditions, or under conditions that create a transient in the speed, load and/or EGR flow, with the transient being repeated continuously to allow collection of ensemble-averaged data that is crankangle-resolved and temporally-resolved during the transient.

Diagnostic

In order to measure the cylinder-to-cylinder distribution of the EGR we are using laser absorption spectroscopy of the CO_2 molecule, a primary component of the recirculated exhaust. An infrared diode laser provides light that is tuned to an absorption transition of CO_2. The output wavelength of the light is modulated so that the absorption transition is continuously wavelength-scanned by the laser. With the extremely narrow output bandwidth

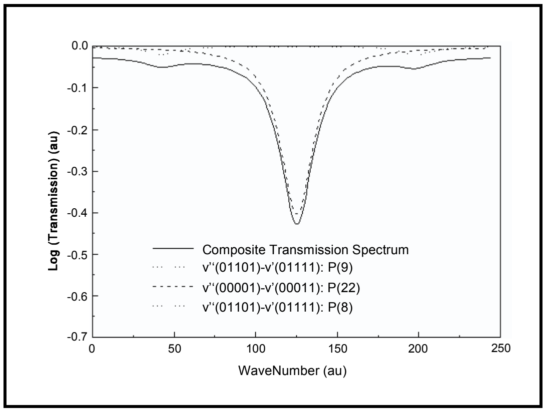

Figure 3. The infrared vibration-rotation spectrum of CO_2 between 2264 cm^{-1} and 2266 cm^{-1}. Also shown is the theoretical composite spectrum which accounts for the effects of the overlapping of the three lines.

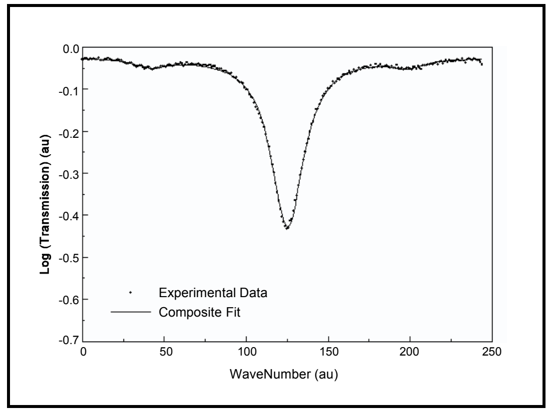

Figure 4. The comparison of a theoretical, non-linear least-squares fit of a Voigt profile with an experimental transmission spectrum measured in the running engine.

of this laser, this procedure allows us to directly measure the absorption-line profile which is a function of the particular species, the concentration of that species, as well as the gas temperature and pressure.

Optical access to the engine is accomplished using a spacer plate between the intake and exhaust manifolds and the cylinder head. The spacer has been designed with windows that provide an absorption path through the intake charge entering each port from the intake manifold. This configuration is illustrated in Figure 1. The laser light is directed through optical fibers to the engine where it is directed through the windows in the spacer plate, passing through the gas entering the intake port. It is then collected in another fiber which directs it to an LN-cooled, In-Sb detector. All of the 'absorption-cell' optics are mounted on a traversing apparatus, as illustrated in Figure 1, which allows the measurements to be made across the intake port of each cylinder.

Data Acquisition and Reduction

A spectral scan through the absorption spectrum of CO_2 is illustrated in Figure 2. This low-pressure (5 torr) scan reveals a strong line at the center of the region bounded by two weaker lines. At manifold pressure and temperature conditions, these lines will be significantly broadened so that the wings of adjacent lines will overlap as illustrated in Figure 3. Thus our measurements will actually yield a composite of the three overlapped lines such as the solid line shown in Figure 3. The composite spectrum shown is the result of the theoretical combination of three Voigt-profile lines.

The data we obtain in our experimental measurements are analyzed by performing a non-linear, least squares fit of the theoretical Voigt profile of the composite spectrum to the experimentally-measured spectrum. The results of this data fitting procedure are used to determine the concentration of CO_2 in the optical path of the laser. An example of this data reduction process is illustrated in Figure 4. The data in Figure 4 were obtained from measurements made in the engine with a significant EGR flow. The excellent fit of these data is a good indication that the data reduction technique has been properly configured. Finally, illustrated in Figure 5 are spectra obtained with the engine running at a low speed and light load for four different levels of EGR flow. These data were fit using

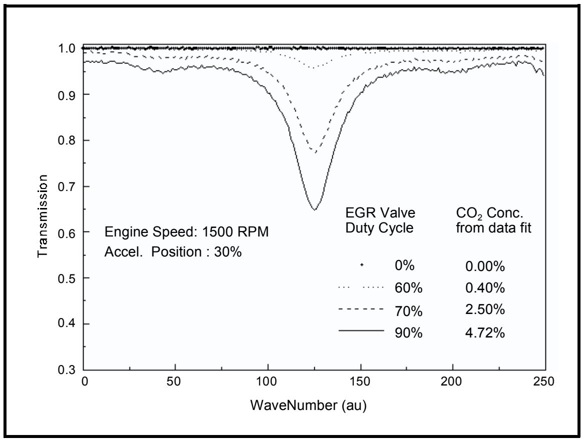

Figure 5. Transmission spectra measured in the engine while running at a low speed and light load for four different EGR flow levels. The results of the data-fit of these spectra are shown in the figure in a listing of CO_2 concentrations.

our data reduction analysis and the results are listed in Figure 5.

Summary

At the present time, we have completed all aspects of the experimental setup. The Volkswagen engine is installed and capable of operation under computer control for both steady and transient conditions. The engine is equipped for optical access, and the IR absorption diagnostic is operational. The analysis required for data reduction has been developed, checked out and is ready. Thus, we are in position to begin acquiring the data needed for the EGR-mixing database. We have identified two suites of computer codes for engine simulation/CFD modeling. We are presently evaluating whether to use one, the other, or even both in the correlation of the data in our experimental database. We don't expect any difficulties having the necessary modeling capabilities available.

List of Acronyms

CFD	Computational Fluid Dynamics
DI	Direct Injection
ECM	Electronic Control Module
EGR	Exhaust Gas Recirculation
TDI	Turbocharged Direct Injection

III.E. Effects of Combustion Air Composition on Light-Duty Diesel Emissions

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Subcontractors
Compact Membrane Systems Inc. (CMS), Wilmington, DE
AutoResearch Laboratories Inc., Harvey, IL

Objectives

- Evaluate intake air nitrogen-enrichment as an alternative to exhaust gas recirculation (EGR) for reduction of oxides of nitrogen (NO_X).
- Evaluate intake air oxygen-enrichment for simultaneous reduction of NO_X and particulate matter (PM).
  - ANL has demonstrated a new O_2-enrichment strategy in a two-cylinder locomotive diesel engine.
  - Verify feasibility in a light-duty compression-ignition direct-injection (CIDI) diesel engine

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 9, Barriers A,B,C

Approach

- Set up and instrument a light-duty CIDI diesel engine for evaluation of performance, emissions, and combustion; obtain baseline maps.
- Evaluate intake air nitrogen-enrichment, with nitrogen supplied by an air separation membrane, for NO_X reduction; compare results to those for operation with and without EGR.
- Evaluate ANL's new approach of utilizing oxygen-enriched intake air for simultaneous reduction of NO_X and PM.

Accomplishments

- Experimental test setup was completed; baseline performance and emissions data were obtained.
- Testing scheme that incorporates prototype membranes was developed for engine experiments with nitrogen-enriched combustion air.
- Prototype air separation membrane units (supplied by CMS) were assembled and installed in the engine intake air system.
- Initial tests with nitrogen-enriched air supplied by prototype membrane units were completed. Additional tests are in progress to evaluate the effects of nitrogen-enriched air on NO_X reductions over a wide range of engine operating conditions.

Future Directions

- Complete tests with N_2-enriching membrane to quantify the benefits and make comparisons with different EGR systems.
- Conduct N_2-enrichment tests with second-generation prototype membrane.
- Develop a prototype membrane for O_2-enrichment tests.
- Quantify the operating strategy for simultaneous reduction of NO_X and PM via O_2-enrichment.
- Demonstrate both O_2- and N_2-enrichment techniques on light-duty vehicles and compare the resulting emissions with PNGV goals.

Introduction

Exhaust gas recirculation (EGR), the most cost-effective way of reducing NO_X emissions from spark-ignition engines, is being considered for use with CIDI diesel engines.

Table 1. Comparison of NEA vs. EGR for NO_X reduction in diesel engines

Nitrogen-Enriched Air	Exhaust Gas Recirculation
• Clean, nitrogen-rich air is delivered by a membrane free of intake airborne PM • No effect on engine life or durability • Acts as a heat exchanger to further lower intake air temperature • Homogeneous mixture makes trade-offs among NO_X, PM, and brake-specific fuel consumption (BSFC) predictable • Requires a membrane separator with 10-12% compensation for flow loss and 1-3 psi for pressure loss	• Unwanted exhaust species in the intake air and an increase in particulates • Presence of sulfur and/or carbon affects engine life and durability • Cylinder-to-cylinder variation is difficult to optimize, and trade-offs among NO_X, PM, and BSFC are unpredictable • Ideally, requires a heat exchanger for cooling and a filter to trap particulates

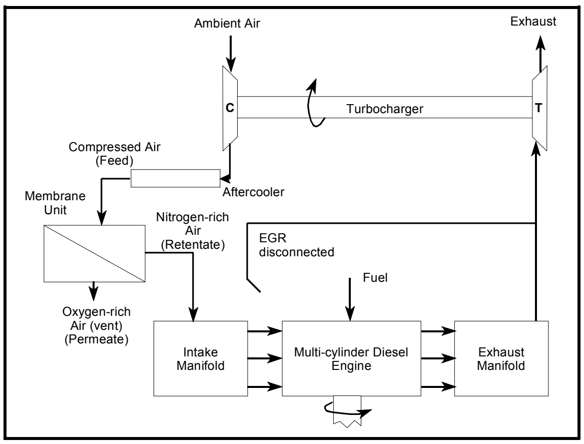

Figure 1. Schematic for supplying NEA in the intake air system via membrane

Figure 2. Membrane units connected at the engine intake air system

However, the applicability of EGR to diesel engines is in doubt because of certain undesirable effects. These include increases in both particulate matter and smoke at higher engine loads, reduced engine durability, and increased cylinder-to-cylinder variations. Argonne has developed a new concept of diluting the intake air with nitrogen-enriched air (NEA) supplied by an air separation membrane. Used as a diluent, NEA can be an effective means of lowering the intake air oxygen concentration, thereby decreasing NO_X formation during combustion. The advantages and requirements of NEA over EGR are compared in Table 1. The program deliverable is to experimentally verify the advantages of NEA over EGR by using prototype membranes on a light-duty CIDI engine.

Argonne has identified an operating regime with oxygen-enriched combustion air whereby simultaneous reduction of PM and NO_x can be achieved. Additional benefits, such as higher gross power, lower peak cylinder pressures, and lower brake specific fuel consumption, were observed. These results were obtained on a two-cylinder locomotive research diesel engine utilizing oxygen from an external source. The second and final deliverable of the present program is to verify that similar benefits are attained by using such a

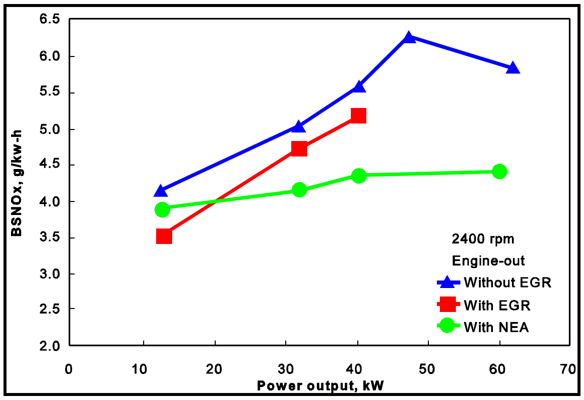

Figure 3. NEA is effective in reducing NO_X at higher boost conditions

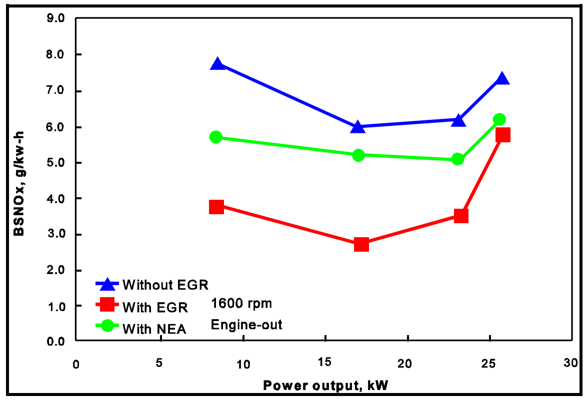

Figure 4. At part loads, NEA surpasses operation without EGR for NO_X reduction

membrane on a light-duty CIDI diesel engine.

Effects of Nitrogen-Enriched Air on NOx Reduction

A 1.9-L turbocharged direct-injection (TDI) diesel engine, rated at 81 kW @4150 rpm, was modified to run on a dynamometer for performance and emissions evaluations. The test engine was originally fitted with EGR and an oxidation catalyst for emissions reduction. Performance and emissions data were obtained at various steady-state engine operating conditions throughout the engine map to evaluate the effects of operating with and without EGR. The engine intake air system was modified to accommodate the nitrogen-enriching membrane. The prototype membrane units (eight modules, each 27 in. long and 3 in. in diameter) were supplied by Compact Membrane Systems, Inc. (CMS), for evaluation on the test engine. Figure 1 shows the schematic arrangement for the membrane supplying NEA, and Figure 2 pictures the membrane units integrated into the engine intake air system.

Test results indicate that at higher boost pressures (>8 psig), the membrane provided nitrogen purity up to 80.0 % from ambient air of 79% by volume. In addition, the membrane lowered the charge temperature by about 10°C. As a result of these dilution and thermal effects combined, NEA gave about 15-20% lower NO_X emissions than did operation with EGR. Figure 3 shows the NO_X emissions from NEA compared with those from operation with and without EGR (constant engine speed of 2400 rpm, various engine loads). For engine operating regimes where boost pressure was less than about 8 psig, NEA was 20-30% better in reducing NO_X emissions than operation without EGR but inferior to operation with EGR (Figure 4). Figure 5 shows NO_X emissions over a wide engine operating range at maximum torque conditions, where EGR was present in the base engine. Evidently, at lower engine speeds and torque conditions, where the boost pressures are not high enough to cause oxygen/nitrogen separation, the membrane provides a modest increase in the nitrogen purity of intake air.

The nitrogen purity and flow depend on membrane design and polymer properties. For this particular prototype membrane, the minimum pressure required at the membrane feed side to provide significant nitrogen-enrichment of ambient air is about 8 psig. The apparent parasitic requirements of the membrane are pressure drop and airflow loss; Figure 6 depicts results from the present tests. However, when the membrane was placed between the after-cooler and the intake manifold, the engine could overcome the pressure drop of about 1-2 psig and flow loss

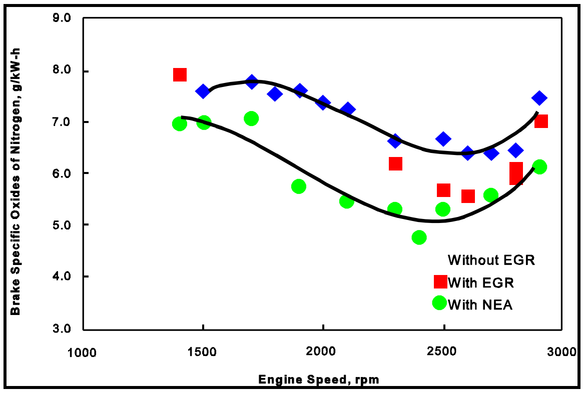

Figure 5. NO_X comparisons at variable speed, maximum torque conditions

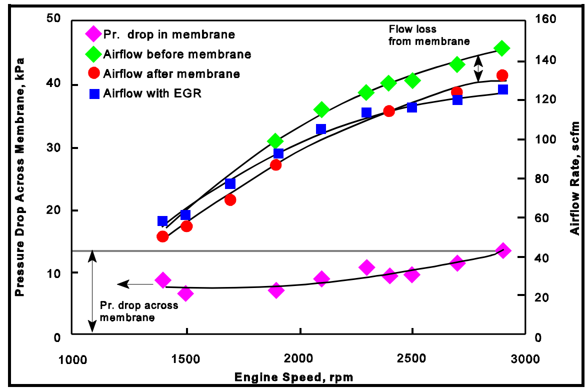

Figure 6. Membrane flow losses are minimal and are compensated for by the turbocharger

of 10-12% of total intake airflow via higher compressor outlet pressures, using exhaust waste gate modulation. As a result, the net brake power was little affected by membrane operation. From the present tests, it is evident that for minimal parasitic requirements, the membrane units need to be matched with the turbocharger for a given engine in the operating range of interest.

Conclusions

On the basis of steady-state engine tests on a 1.9-L TDI diesel engine using nitrogen-enriched combustion air supplied by prototype air separation membrane units, the following observations were made:

- Dilution and thermal effects with NEA yield lower NO_X emissions - 10-20% reduction at part loads and lower speeds and 20-30% reduction at higher loads and/or at higher speeds, compared to operation without EGR.
- At higher (>~8 psig) boost conditions, NEA is 15-20% better than EGR in reducing NO_X emissions.
- Without optimizing membrane or engine systems, and within the range of test conditions examined, NEA has no detrimental effect on performance and provides NO_X reduction similar to or better than that with EGR.
- Membranes can be matched to obtain greater NO_X reductions in the engine operating range of interest.
- The flow and pressure losses from the membrane are low and can be fully compensated for with proper waste gate modulation.

Publications

Poola, R., et al., "Nitrogen-Enriched Air as an Alternative to EGR for NO_X Reduction in Diesel Engines," Paper communicated to the SAE International Congress and Exposition, March 2000.

List of Acronyms

ANL	Argonne National Laboratory
CIDI	Compression Ignition Direct Injection
CMS	Compact Membrane Systems, Inc.
EGR	Exhaust Gas Recirculation
NEA	Nitrogen-Enriched Air
PM	Particulate Matter
TDI	Turbocharged Direct Injection

III.F. Diesel Hydrocarbon Speciation

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Objectives

- The objective of the proposed effort is to determine chemical composition of the HC species in the exhaust as a function of post-injection conditions.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 1A, Barriers A,B

Approach

- Previous bench studies with catalyst materials have shown that diesel fuel does not contain the ideal mix of HC for reducing NO_X species. The heavy HC species in diesel are often blamed for the poor performance of lean NO_X catalyst materials in actual engine tests. By injecting the fuel in the cylinder during the exhaust stroke, the HC mix will likely change to lower molecular weight species that may increase the NO_X reduction of the catalyst. Timing and placement of the post-injection process will affect what HC species the catalytic converter sees. ORNL's task is to perform the engine tests and speciate the HCs and determine what is required by the catalytic emissions control unit for NO_X reduction. Two engines as well as different catalytic emissions control units are planned for investigation.

Accomplishments

- All of the testing has been completed on the Navistar engine. Data analysis is 90% complete. The installation of the Series 50 will be completed July 30, 1999.

Future Directions

- Possible investigation of different types of catalytic emission control technologies.

Introduction

This CRADA is concerned with the role different HC species play in the reduction of NO_X on catalyst materials. Two engines are being used: a medium duty 1994 Navistar T444E 7.3 L turbocharged, direct-injected engine, and a heavy duty 1998 DDC Series 50

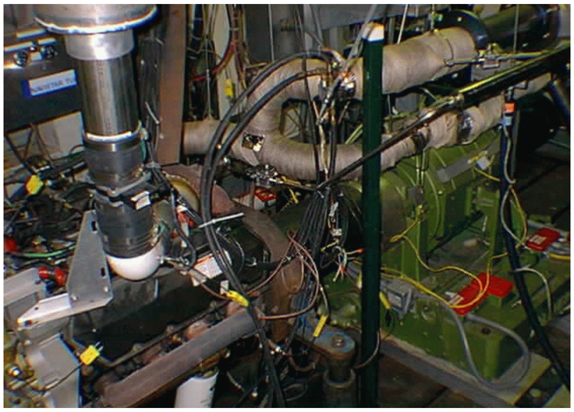

Figure 1. Navistar Engine Laboratory Setup

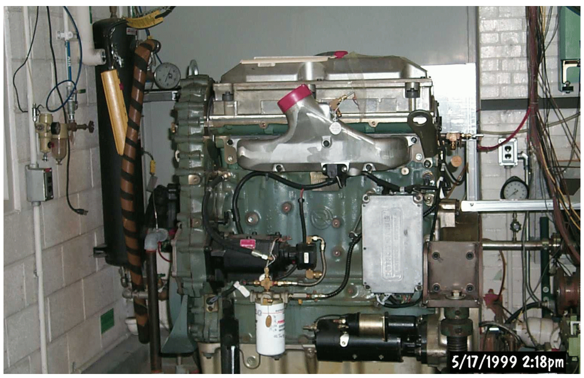

Figure 2. DDC Series 50 Engine Setup in Progress

8.3 L engine. The Navistar engine is equipped with in-cylinder injection and in-pipe injection and is shown installed on the test cell in Figure 1. After work was completed in April 1999 on the Navistar engine, the Series 50 engine was installed on the same location. The Series 50 is shown during installation in Figure 2.

The test matrix on the Navistar engine was completed for different in-cylinder HC injection rates and injection crank angles, and included more than 80 operating points. It was determined that in-cylinder HC injection affects HC species. Species were affected by injection crank angle and the amount of HC injected and total PM increased as injection rates increased. Regulated emissions as well as PM SOF, PM SO_4, bag HCs, aldehydes and ketones, and semivolatile HCs were determined.

Effect of HC Injection Method on HC Speciation

More than 40 HC species were identified. Quantitation for more than 40 species and more than 100 others observed. Some measurement issues include wide dynamic range observed between no HC injection and HC injection. For instance, the GC/MS is unable to measure engine out (no injection) HCs except at high speed/low load due to low overall HC. However, at moderate to high HC injection rates, there were very high levels of HCs. Furthermore, HC carryover in the sampling system became an issue.

Summary

This CRADA sets out to understand the effects of HC injection on the HC species in the exhaust. The objective is to further the understanding of HC utilization by lean NO_X catalytic emissions control systems.

III.G. HCCI Combustion Fundamentals Using In-Cylinder Diagnostics

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Objectives

- Provide the fundamental understanding (science base) necessary to overcome the obstacles limiting the development of a practical HCCI (homogeneous charge compression ignition) engine.
  - HCCI engines can overcome all three technical barriers to the current diesel engine.
  - This is a new project for FY99 that is planned to continue for several years.
- Transfer results to industry.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 4, Barriers A,B

Approach

- Establish a laboratory to investigate HCCI combustion.
  - All-metal engine to establish operating points and develop the combustion-control strategies necessary to maintain proper combustion across the load-speed map.
  - Optically accessible engine to apply advanced laser diagnostics to investigate the details of HCCI combustion, e.g. fuel/air/residual mixture preparation, charge stratification, combustion quench at walls, and sources of hydrocarbon emissions.
- Chemical-kinetic rate calculations of HCCI to guide engine design and operating-point selection.
  - Results will be compared with engine data when the engines come on line.

Accomplishments

- Laboratory setup is on track as planned for FY99.
  - Design of laboratory has been completed, and existing experiments have been removed.
  - Designs for modification of the laboratory and support facilities have been completed and work is underway.
  - Long lead time capital items (air compressor and double-ended dynamometer) have been specified and ordered.
- Engine acquisition and modification is on track as planned for FY99.
  - Base engine has been selected and cylinder-head modifications are underway.
  - Variable valve-timing (VVT) systems have been researched and a system selected.
  - Metal engine and dynamometer expected to be in place near the end of FY99.
- CHEMKIN (chemical-kinetics rate code) operation has been established and computations have been made across much of the parameter space.
  - Calculations appear to capture the trends correctly, and should be a valuable design tool.
  - This modeling has already provided new insight into engine design and fuel-type selection.

Future Directions

- Complete the installation of the metal engine, including modifications for balanced single-cylinder operation and the design, fabrication, and installation of engine subsystems.
  - Obtain VVT system if money permits.

- Shakedown testing of metal engine - expected in late FY00.
- Establish an initial smooth-firing operating point. Then, begin testing a range of operating parameters about this point.
  - Compare results against CHEMKIN calculations and determine predictive capabilities.
- Install second base engine and design the modifications for optical access and rapid cleaning.
  - Expected to be operational in late FY01, at which time in-cylinder studies will commence.
- Conduct CHEMKIN calculations for a wider range of conditions and in support of metal engine testing, as engine becomes operational.

Background and Objectives

Homogeneous charge compression ignition (HCCI) is an alternative engine combustion process that can provide high, diesel-like efficiencies, while producing very low NO_X and particulate emissions. There is also a high probability that HCCI engines would be less expensive than diesels since they would use lower pressure fuel-injection equipment. Accordingly, HCCI has the potential to overcome all three of the technical barriers to the high-speed diesel engine listed by PNGV, i.e. NO_X emissions, particulate emissions, and cost.

The advantages of HCCI are even more significant considering our relatively complete understanding of diesel-engine combustion and emissions. Advanced laser diagnostics have shown the controlling physics, and they indicate that soot (particulates) might be further reduced, but that substantial NO_X reductions are unlikely beyond those achievable with EGR (exhaust gas recirculation) and more traditional methods. Since the NO_X-reducing potential of these technologies have been extensively tested, this means that diesel engines will not meet 2004 Tier 2 passenger-car NO_X standards without significant aftertreatment.

However, NO_X aftertreatment technologies for diesels are still inadequate, despite extensive efforts. The best aftertreatment technologies, combined with maximum in-cylinder reduction using EGR, might approach the 2004 passenger-car standard, but a major technological breakthrough would be required to meet any more-stringent standard. Moreover, the EGR and aftertreatment technologies are likely to be complex, expensive, and entail a fuel consumption penalty.

HCCI eliminates NO_X and particulate emissions at their source by operating in a homogeneous, lean or dilute (using EGR) mode. This process has been shown to produce high efficiencies with extremely low NO_X and particulate emissions. However, there are challenges to implementing HCCI in a practical engine, including: controlling the combustion timing and heat release rate across the load-speed map, transients, and unburned hydrocarbon emissions. Although these issues require a serious research effort, the obstacles may well be less difficult to overcome than those of the diesel.

The objective of the current project is to develop the fundamental understanding necessary to overcome the problems challenging HCCI. This research will be conducted in close cooperation with both the automotive and heavy-duty diesel industries, most of whose members have expressed a strong interest in HCCI. Results will be presented at the cross-cut diesel CRADA meetings, and input from industry will be incorporated into the work plans.

Laboratory and Engines

An HCCI engine combustion laboratory is being established that will contain two engines of the same basic design. First, an all-metal engine will be used to establish operating points and develop combustion-control strategies. Second, an optically accessible engine will be used to apply advanced laser diagnostics to the in-cylinder processes. Examples of needed studies include: investigations of the fuel/air/residual mixture preparation using various fuel-injection and residual-mixing techniques; the effects of small inhomogeneities and global stratification; combustion quench at the walls; and other sources of unburned hydrocarbons.

Since HCCI combustion is determined by the chemical-kinetic rates, several features for controlling these rates are being designed into the research engines. The main features include:

- Multiple fueling systems: 1) a fully premixed system with heater to insure complete fuel vaporization; 2) direct in-cylinder injection to allow variable fuel-compression time and/or controlled stratification.
- Flexible fuel capability: The fueling system will accommodate diesel-like, gasoline-like, and gaseous fuels since the best fuel for HCCI is a current research topic.
- Variable valve timing (VVT): A computer-controlled, electro-hydraulic VVT system will allow the effective compression ratio and the amount of hot residuals remaining in the cylinder to be varied dynamically as the engine is running.
- Intake charge tailoring: To investigate turbocharging and the use of hot EGR for control of combustion timing, the intake system will be equipped with an air compressor, an electrical heater, and the ability to add simulated exhaust gases.
- Variable-swirl porting: The engine will have both a quiescent and a swirl intake port, allowing swirl ratios from near zero to approximately three.

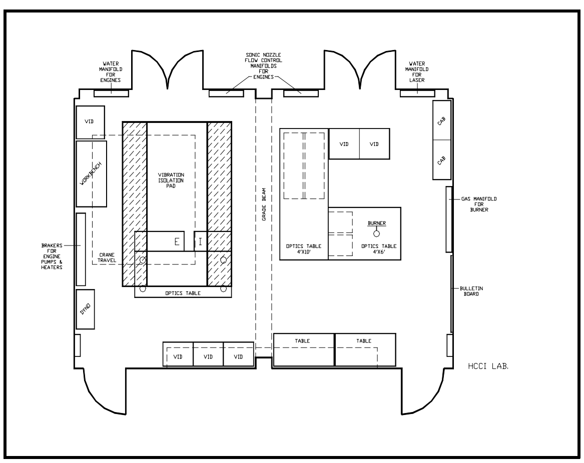

Figure 1. Layout of the Sandia HCCI Engine Combustion Laboratory

Several base engines were researched, and the Cummins B-series has been selected. This SUV-sized (0.98 l/cyl.) engine is capable of speeds above 3500 rpm, and can be readily converted to the required single-cylinder operation. Cummins will provide two engines with modified intake porting to meet our variable-swirl requirements at no charge (saving > $100,000). A production head and mechanical drawings have been received, and the engine modifications are underway.

The laboratory-layout design has been completed and is shown in Figure 1. The two engines will be mounted on the vibration isolation pad at opposite ends of a double-ended dynamometer. To create a laboratory of sufficient size, two adjacent rooms are being combined. Existing experiments and the wall between the rooms have been removed, and all other demolition work has been completed. The construction drawings for the facility modifications (isolation pad, air-compressor installation, electrical and mechanical systems, etc.) are complete. We expect the construction

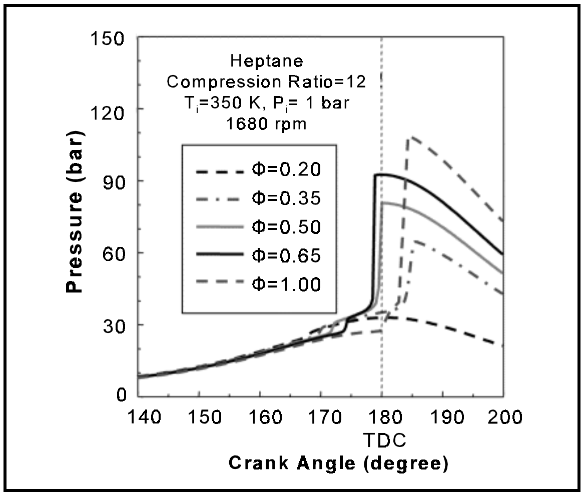

Figure 2. CHEMKIN Calculations Showing the Effect of Equivalence Ratio on HCCI Engine Combustion for N-heptane Fuel (cetane no. 56)

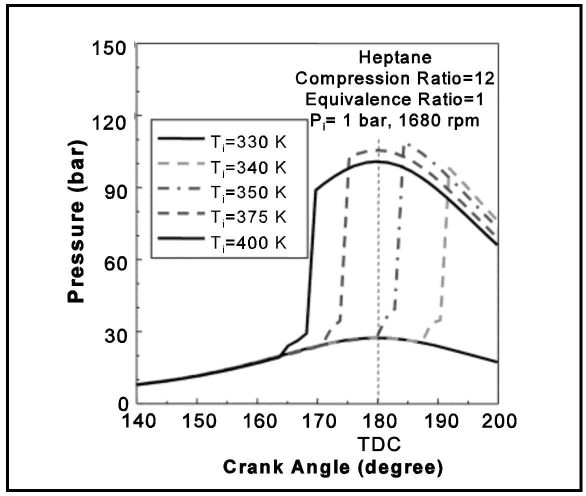

Figure 3. CHEMKIN Calculations Showing the Effect of Intake Temperature on HCCI Engine Combustion for N-heptane Fuel (cetane no. 56)

contract to be signed shortly, and the work to be completed by mid-September 1999.

Chemical-Kinetic Rate Computations

As discussed above, HCCI combustion is controlled by the chemical-kinetic reaction rates, which are affected by a variety of parameters, including: temperature, mixture-composition, fuel-type, compression-ratio, and speed. Kinetics-rate computations will be used to facilitate the selection of engine-design parameters and operating points within this large parameter space. Operation of the CHEMKIN code has been established with the full chemistry for n-heptane and iso-octane from Lawrence Livermore National Laboratory. These fuels span the cetane/octane range of interest, and calculations of HCCI have been run for several conditions.

Figures 2 and 3 present examples of CHEMKIN results for n-heptane (cetane no. 56) which is a good chemical-kinetic surrogate for diesel fuel. As shown in Figure 2, variation of the fuel/air mixture causes two competing effects on the HCCI ignition timing. Lower equivalence ratios have a larger ratio of specific heats, so the engine compresses the mixture to a higher temperature, causing the initial combustion to occur earlier. However, higher equivalence ratios have faster "low-temperature" chemistry, so the delay between the initial and main combustion is shorter. Due to competition between these two effects, the fastest main ignition occurs with an equivalence ratio of 0.65. Although the data in Figure 2 show that variations in the fuel/air mixture cause the ignition timing to shift by several degrees, Figure 3 shows that variations of this magnitude are well within the range that can be compensated for by changing the intake temperature with hot EGR.

Summary and Future Plans

Significant progress has been made toward establishing an HCCI engine laboratory. The layout design, construction drawings, and demolition work have been completed, and the facilities construction work is on track to be completed by mid September.

The Cummins B-series has been selected as the base engine. Intake-valve port modifications are underway, and engine delivery is expected within the next month. VVT systems have been researched, and a programmable, electro-hydraulic system has been selected.

Operation of the CHEMKIN code with the full chemistry for n-heptane and iso-octane (from LLNL) has been established, and computations of HCCI have been run for several conditions.

The laboratory setup and CHEMKIN calculations will continue in FY00 as noted in the bulleted outline above. CHEMKIN results will be compared with data from the all-metal engine when it becomes operational in late FY00 or early FY01. The components for the optical engine will be designed in FY00, and the engine is expected to be operational near the end of FY01 at which time optical measurements will begin.

Presentations

J. Dec, "HCCI Combustion: Plans and Progress," PNGV, 4SDI Tech Team Meeting, Jan. 14, 1999.

J. Dec and P. Kelly-Zion, "HCCI Combustion: Plans and Progress," Cross-Cut Diesel CRADA Meeting, Feb. 3-4, 1999.

J. Dec and P. Kelly-Zion, DOE Review Meeting, Washington, D.C., April 8, 1999.

J. Dec and P. Kelly-Zion, Seminar on the HCCI project at Delphi Automotive, May 5, 1999.

J. Dec and P. Kelly-Zion, "HCCI Engine Combustion Fundamentals Using In-Cylinder Diagnostics," DOE Diesel Combustion Review, June 21-23, 1999.

Acronyms

PNGV	Partners for a New Generation Vehicle
HCCI	Homogeneous Charge Compression Ignition
EGR	Exhaust Gas Recirculation
VVT	Variable Valve Timing
FY	Fiscal Year
PM	Particulate Matter
VVT	Variable Valve Timing
SUV	Sport Utility Vehicle
LLNL	Lawrence Livermore National Laboratory

III.H. Computational Hydrodynamics for Advanced Design (CHAD) Modeling Activities

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Objectives

- Develop a version of the CHAD code for diesel engine simulations. CHAD is a next-generation parallel hydrodynamics code beyond the KIVA code.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 4, Barriers A,B

Approach

- Install the KIVA spray model in CHAD, upgrade the spray model with improvements suggested by recent research, and validate the model in comparisons with experimental data of Sandia National Laboratories (SNL).
- Develop a meshing strategy for performing full-cycle simulations with CHAD. Perform full-cycle simulations of experimental diesel engines at SNL.
- Assist industry researchers in refining the CHAD diesel model by comparing with experiments in realistic engine geometries.

Accomplishments

- A parallel version of the KIVA spray model was installed in CHAD. We began incorporating a higher order particle/grid interpolation scheme, which will improve the numerical accuracy of the spray calculation. A spectral model was developed to predict drop sizes formed in primary spray jet breakup.
- A proposed meshing strategy for full-cycle simulations, based on global mesh changes, was tested on a generic 2-valve engine geometry. The conclusion of this study was that the proposed strategy must be modified to allow for local mesh connectivity changes. A study was also initiated of the importance of using hybrid meshes, as opposed to pure tetrahedral grids, in engine calculations.
- Caterpillar and Los Alamos have performed steady port flow calculations using CHAD. The results show that CHAD is more accurate than some commercial computational fluid dynamic (CFD) codes, but that CHAD is also slower. For a steady port flow calculation CHAD demonstrated parallel scaling performance using up to 64 processors of an SGI Origin 2000.

Future Directions

- Complete installation of the higher order particle/grid interpolation in the CHAD spray model and compare with SNL data.
- To perform full cycle engine simulations, local mesh connectivity change strategies, such as local remesh and remap, reconnection, and adding and merging nodes will be explored. Further comparisons will be performed of solution accuracy using tetrahedral and hybrid grids.
- Install a Newton-Krylov solver in CHAD to attempt to improve computational speed.
- Continue to provide support to industrial users of CHAD.

Introduction

CHAD is a next-generation hydrodynamics code for combustion applications that is highly parallel and portable, that utilizes hybrid-unstructured grids for geometric flexibility, and that has a variable explicit/implicit difference scheme for computational efficiency. Our recent efforts have focused on meshing strategies for diesel engine calculations and on modeling steady port flow with researchers at Caterpillar Corporation.

Meshing Strategies for Diesel Engine Calculations

A full-cycle meshing capability is needed before CHAD diesel engine calculations can be performed. Full-cycle meshing of a generic 2-valve engine has been accomplished and has revealed some deficiencies, and suggested some improvements, in a proposed engine meshing strategy. The proposed meshing strategy relies on mesh smoothing with occasional global remappings. The mesh smoothing is performed as the mesh moves to follow valve and piston motion. The purpose of this smoothing is to maintain mesh quality without changing mesh topology. When mesh quality can no longer be maintained, a new, better quality mesh is generated, and the computed solution on the old mesh is remapped onto the new mesh. The number of meshes should be kept small because each remapping introduces some numerical inaccuracy into an engine calculation.

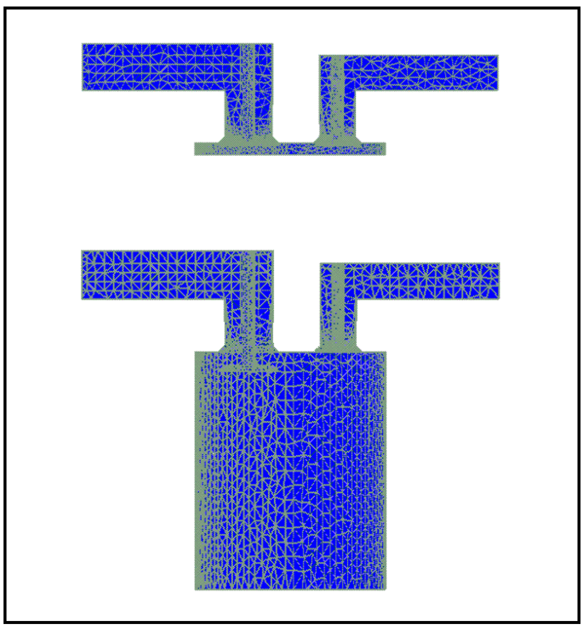

Figure 1. Surface mesh of generic 2-valve engine at 0° (top) and 180° (bottom) ATDC intake stroke

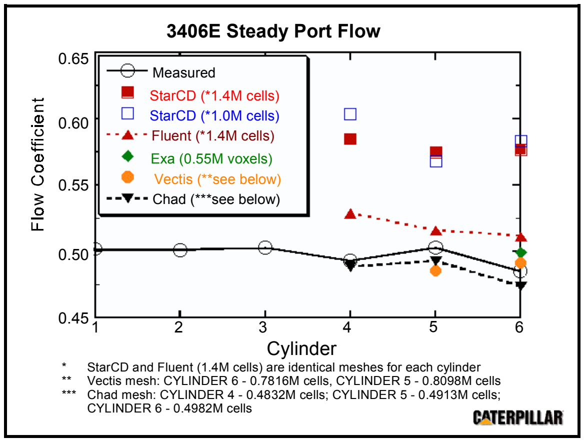

Figure 2. Flow coefficient variation versus cylinder for 3406E port flow

Figure 1 shows the surface mesh in the 2-valve engine mesh at two different crank angles. Mesh generation was performed with the ICEM-CFD™ mesh generator and was based on the use of tetrahedra. Mesh smoothing used a combination of elastic body motion and “smart” Laplacian smoothing. LANL found that even in this relatively simple engine geometry, 14 topologically different meshes were needed to maintain good mesh quality for a complete 720° engine cycle. LANL is currently looking at methods for reducing this unacceptably large number of meshes.

Steady Port Flow Calculations

Steady port flow calculations performed by Caterpillar, with help from Los Alamos, showed the high accuracy of the CHAD code for simulating flows in complex geometries.

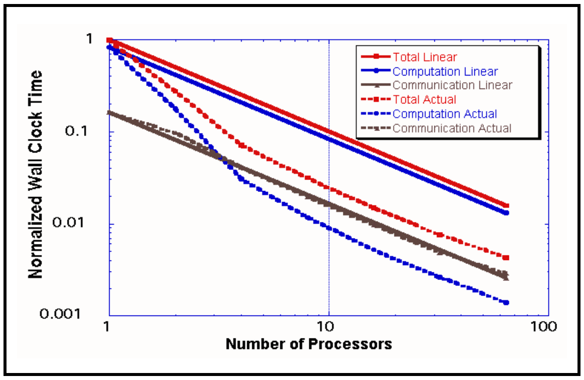

Figure 3. Computational time (normalized by time for 1-processor calculation) versus number of processors for calculation of SNL small-bore engine

Figure 2 shows the results of CHAD calculations compared with those of some commercial codes and with experiments. Each of three experiments measured the flow coefficient through an intake manifold and one cylinder, with the flow through the other cylinders blocked. CHAD was the only code that reproduced the observed variation in flow coefficient versus cylinder.

CHAD also demonstrated parallel scaling performance in steady flow calculations of flow through one intake port and cylinder of an SNL small-bore diesel engine. The calculations were performed on an SGI ORIGIN 2000 at Los Alamos. As shown in Figure 3, the computational time scaling was actually super-linear up to 4 processors.

References/Publications

P.J. O'Rourke and M.S. Sahota, "A Variable Explicit/Implicit Method for Calculating Advection on Unstructured Meshes," J. Comput. Phys. 143, 312 (1998).

P.J. O'Rourke and M.S. Sahota, " NO-UTOPIA: A Parallel, Implicit Method for Calculating Three Dimensional Fluid Flows on Unstructured Grids, " Los Alamos National Laboratory Report (in preparation).

S. Subramaniam and P.J. O'Rourke, "Preliminary Computations of Diesel Sprays: Numerical Convergence and Implications for Modeling," (submitted to Atomization and Sprays).

S. Subramaniam, "A Spectral Model for Primary Breakup of Round Turbulent Liquid Jets," Los Alamos National Laboratory Report (in preparation).

P.J. O'Rourke, M.S. Sahota, and S. Zhang, "A Parallel, Unstructured-Mesh Methodology for Device-Scale Combustion Calculations," in special issue Multidimensional Simulation of Engine Internal Flows of Revue de l'Institut Francais du Petrole, edited by T. Baritaud, Vol. 54, pp.169-174 (1999).

Information on the ICEM-CFD grid generation software can be found on the internet at www.icemcfd.com.

List of Acronyms

CFD	Computational Fluid Dynamics
CHAD	Computational Hydrodynamics for Advanced Design
LANL	Los Alamos National Laboratory
SNL	Sandia National Laboratory

IV. EXHAUST GAS NO_X EMISSION CONTROL R&D

IV.A. Plasma-Assisted Catalytic Reduction for Automotive Diesel Engines

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Objectives

- Understand how the efficiency of the Plasma-Assisted Catalytic Reduction (PACR) process scales from heavy-duty to light-duty automotive diesel engine exhaust.
- Characterize the performance of the PACR process when the hydrocarbon additive is diesel fuel instead of propene.
- Design and fabricate full-scale prototype of a PACR processor capable of treating the exhaust from a 50 kW diesel engine.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 1B, Barriers A,B

Approach

- Use simulated diesel exhaust to measure the NO_X reduction efficiency as a function of initial NO_X and hydrocarbon concentration, electrical power input to the plasma, and water vapor level in the exhaust.
- Install means for injecting diesel fuel into the PACR processor.
- Use slipstream of exhaust from Cummins diesel engine to measure the NO_X reduction efficiency as a function of diesel fuel concentration in the exhaust and electrical power input to the plasma.

Accomplishments

- Established how the efficiency of the PACR process scales from heavy-duty to light-duty automotive diesel engine exhaust.
- Built and tested a system for injecting diesel fuel into the PACR processor.
- Demonstrated feasibility of the PACR process on a Cummins B5.9 diesel engine exhaust, using diesel fuel as the hydrocarbon additive.
- Completed design and assembly of the full-scale PACR processor unit.

Future Directions

- Evaluate and refine full-scale prototype of the PACR processor.
- Test long-term durability of the NO_X reduction catalyst against sulfur and soot.
- Test long-term durability of the plasma processor against insulator flashover.
- Optimize timing of diesel fuel injection into the PACR processor for transient operations.
- Assemble full-scale PACR system in mobile support frame in preparation for deployment in a vehicle.

Introduction

Plasma-assisted catalytic reduction (PACR) is based on the selective plasma oxidation of NO to NO_2, followed by the selective catalytic reduction of NO_2 to N_2 (see Figure 1). Addition of hydrocarbon to the exhaust is necessary to efficiently accomplish both the plasma oxidation to NO_2 and the catalytic reduction to N_2. The PACR process has been tested on heavy-duty diesel engine exhaust, using propene as the hydrocarbon additive. The first objective of this work is to understand how the PACR process efficiency scales when applied to light-duty automotive diesel engine exhaust. The second objective is to characterize the performance of the PACR process when the hydrocarbon additive is diesel fuel instead of propene.

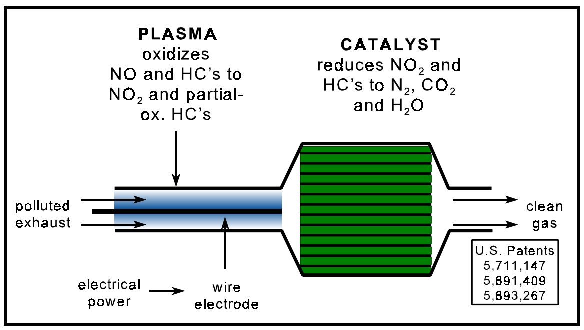

Figure 1. Schematic of the plasma-assisted catalytic reduction (PACR) process

Results

Figure 2 shows the plasma oxidation efficiency in a gas mixture simulating a heavy-duty diesel engine exhaust, with propene as the hydrocarbon additive. The oxidation efficiency is proportional to both the electrical energy density applied to the plasma and the amount of hydrocarbon added to the exhaust. With 600 ppm initial NO, the plasma requires an electrical energy density of 30 J/L to achieve maximum oxidation of NO to NO_2 when $C_1/NO_x = 6$ (i.e., 1200 ppm of C_3H_6). For a steady engine load, 30 J/L corresponds to around 3% of the engine power output.

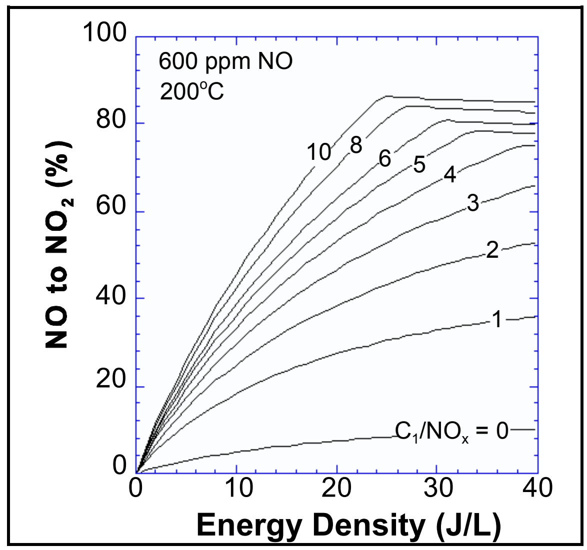

Figure 2. Plasma oxidation of NO to NO_2 in a gas mixture simulating a heavy-duty diesel exhaust, using propene additive

Figure 3 shows how the electrical power requirement in the plasma changes when applied to a gas mixture simulating light-duty

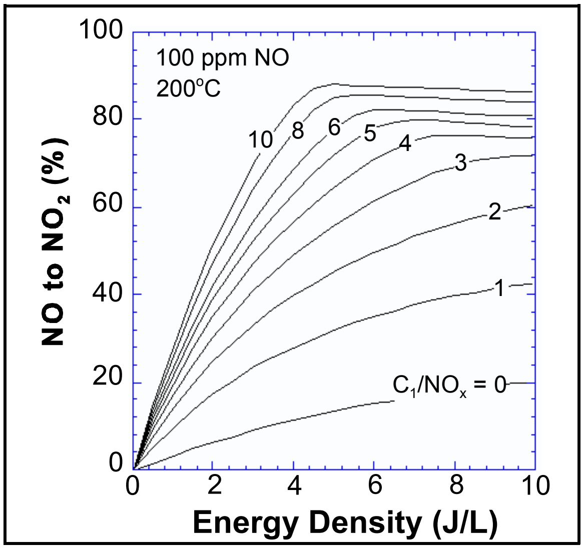

Figure 3. Plasma oxidation of NO to NO_2 in a gas mixture simulating a light-duty diesel exhaust, using propene additive

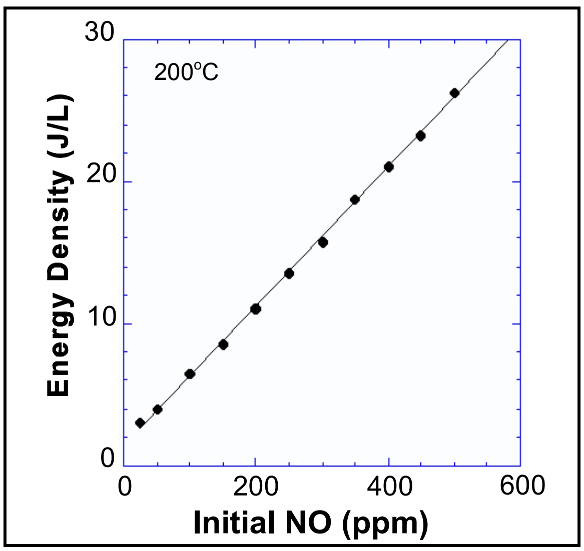

Figure 4. Electrical energy density required by the plasma to achieve maximum oxidation of NO to NO_2 with $C_1/NO_X = 6$

engine exhaust. With 100 ppm initial NO, the plasma requires only 6 J/L to achieve maximum oxidation for the same $C_1/NO_x = 6$ (i.e., 200 ppm of C_3H_6). As shown in Figure 4, the electrical power requirement in the plasma is linearly proportional to the initial NO concentration. Plasma oxidation in a light-duty

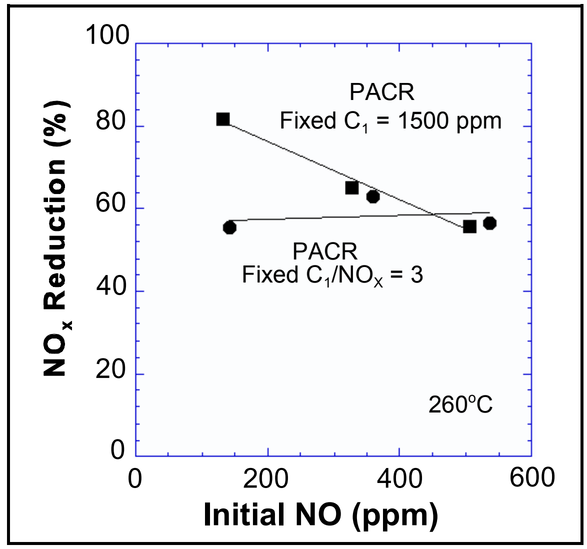

Figure 5. NO_X reduction by PACR as a function of initial NO concentration

diesel engine exhaust is much easier because of the lower initial NO_X level.

The number of hydrocarbon molecules determines the number of catalyst sites on which NO_X could be reduced. As shown in Figure 5, it is necessary to maintain the same number of hydrocarbons, rather than the same hydrocarbon/NO_X ratio, in order to reduce approximately the same number of NO_X molecules over the catalyst. The light-duty application will therefore require a higher hydrocarbon/NO_X ratio to reduce the same number of NO_X.

Adsorption of both the NO_2 and the hydrocarbon on the catalyst is required to accomplish the reduction of NO_X. Water vapor in the exhaust degrades the NO_X reduction efficiency because it inhibits the adsorption of NO_2 on the catalyst. As shown in Figure 6, it is difficult to achieve high NO_X reduction efficiency in a light-duty engine exhaust with 5% or more water vapor, even if the hydrocarbon level is very high. In the presence of 5% or more water vapor, the PACR process will have the same NO_X reduction efficiency for light-duty and heavy-duty applications.

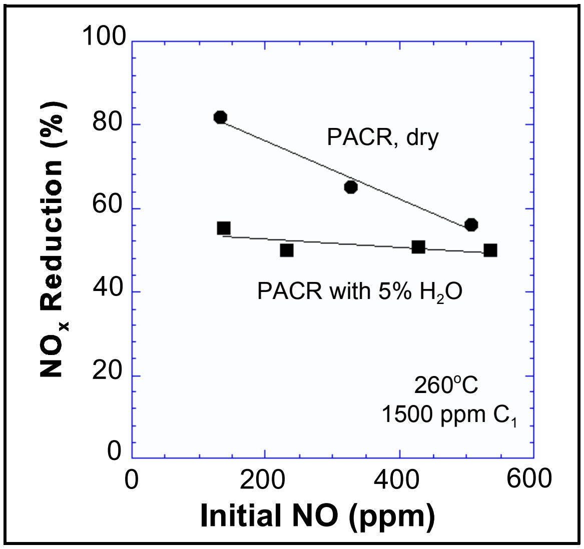

Figure 6. Effect of water vapor on the NO_X reduction by PACR

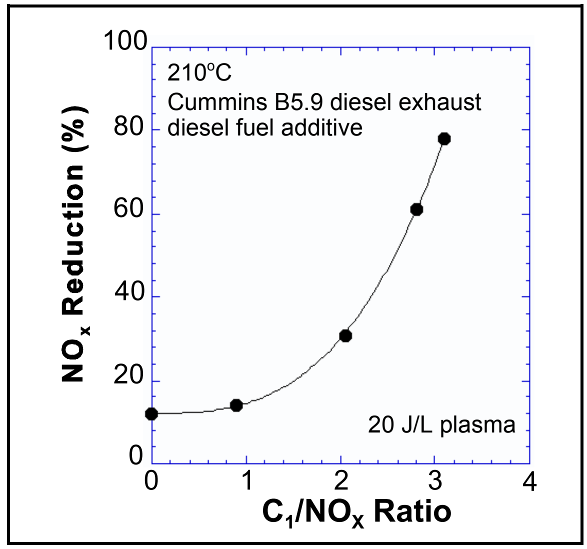

Figure 8. NO_X reduction efficiency by PACR, using diesel fuel as a hydrocarbon additive

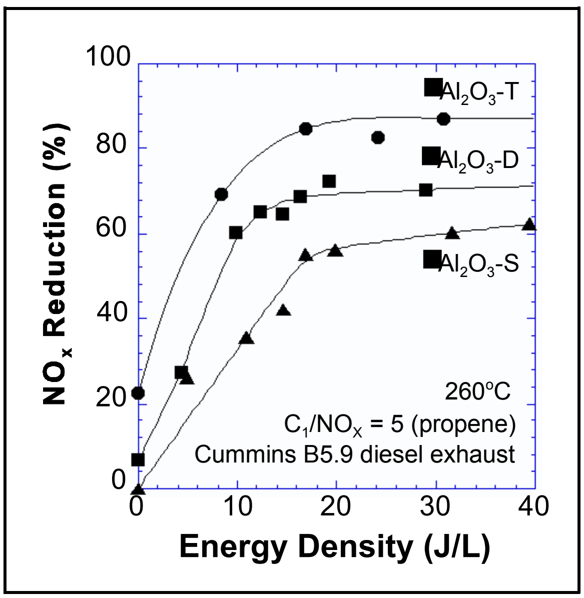

Figure 7. NO_X reduction efficiency by PACR for three different formulations of γ-Al_2O_3

To achieve a high NO_X reduction efficiency, it is necessary to improve the basic properties of the catalyst itself. Figure 7 shows the NO_X reduction efficiency by PACR for three different formulations of a γ-Al_2O_3 catalyst. We have performed similar tests on various zeolite catalysts.

A system for injecting diesel fuel into the PACR processor was built and tested. Figure 8 shows the NO_X reduction efficiency by PACR, using diesel fuel as hydrocarbon additive, in a heavy-duty diesel engine exhaust. High NO_X reduction efficiency can be achieved at a fairly modest C_1/NOx ratio. Obtaining the same high NO_X reduction efficiency in a light-duty diesel engine exhaust will require a much higher C_1/NO_X ratio, as we have learned from the scaling study in Figure 6.

Conclusions

Treatment of light-duty diesel engine exhaust is expected to be more difficult compared to that of heavy-duty engines because of the lower initial-NO_X concentration (100 ppm instead of 600 ppm), lower exhaust temperature (200°C instead of 400°C), and higher NO_X reduction requirement (over 90% instead of 50%). We have established how the efficiency of the PACR process scales from heavy-duty to light-duty automotive diesel engine exhaust. Plasma oxidation in a light-duty application requires much less electrical power because of the lower initial-NO_X level and lower exhaust flow rate. The water vapor in the exhaust is a serious limiting factor in getting high NO_X reduction efficiency on the catalyst at low initial NO_X concentrations. The total fuel penalty for the PACR process in a light-duty application is determined mostly by the amount of hydrocarbon additive required by the catalyst. At low temperatures, the plasma can significantly enhance the catalytic reduction of NO_X, with little fuel penalty incurred in the plasma process.

Publications/Presentations

B.M. Penetrante, "Feasibility of Plasma Aftertreatment for Simultaneous Control of NO_X and Particulates", presented at the 1999 Diesel Engine Emissions Reduction Workshop, July 5-8, 1999, Castine, Maine.

B. M. Penetrante, "Plasma-Assisted Catalytic Reduction of NO_X Over Zeolite Catalysts", presented at the 1999 Diesel Engine Emissions Reduction Workshop, July 5-8, 1999, Castine, Maine.

B.M. Penetrante, "Limiting Factors and Effect of Sulfur on Plasma-Assisted Catalytic Reduction of NO_X in Light-Duty Vehicle Exhaust", presented at the 1999 Diesel Engine Emissions Reduction Workshop, July 5-8, 1999, Castine, Maine.

B.M. Penetrante, R.M. Brusasco, B.T. Merritt, W.J. Pitz, G.E. Vogtlin, M.C. Kung, H.H. Kung, C.Z. Wan, K.E. Voss, "Plasma-Assisted Catalytic Reduction of NO_X in Lean-Burn Engine Exhausts", presented at the 16th North American Catalysis Society Meeting, May 30 - June 4, 1999, Boston, MA.

List of Acronyms

PACR	plasma-assisted catalytic reduction
SCR	Selective catalytic reduction
ppm	parts per million
J/L	joules per standard liter

IV.B. Plasma-Assisted Catalysts

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Objective

- Develop an aftertreatment system that will achieve 90% NOx reduction using less than 5% of the engine power on a compression ignition direct injection (CIDI) diesel-fueled engine. The program supports goals of the Partnership for the Next Generation of Vehicles (PNGV).

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 1B, Barriers A,B

Approach

- A non-thermal plasma in conjunction with new catalytic materials is being developed to reduce NO_X emissions with a secondary goal of oxidizing hydrocarbons and particulate. A partnership between the Pacific Northwest National Laboratory and the Low Emissions Technologies Research and Development Partnership (LEP) consisting of Ford, General Motors and DaimlerChrysler has been established under a Cooperative Research and Development Agreement (CRADA). In addition, Oak Ridge National Laboratory (ORNL) is collaborating with PNNL and the LEP in the area of ceramic synthesis and engine testing.

Accomplishments

- New catalysts have been discovered that can reduce NO_X when placed in or down-stream from a plasma reactor. Bench tests with simulated diesel exhaust show that a plasma-catalyst system can reduce up to 70% of NO_X emissions at temperatures typical of CIDI exhaust (150-370°C) and an equivalent fuel penalty of 1%.
- Tests on a slip-stream of diesel engine exhaust from a diesel generator show that 53% NO_X reduction can be achieved at 200°C and an equivalent fuel penalty of 6%.
- SO_2 in simulated mix does not degrade catalyst activity.
- A full-scale prototype plasma reactor/catalyst system has been fabricated and delivered for testing. Tests on a VW 1.9L TDI diesel engine are ongoing.
- Nitrogen balance has been obtained for the best catalysts.
- The role of NO_2 over the best catalysts has been determined.

Future Directions

- Develop higher activity catalysts that are durable over long times in a real diesel environment.
- Develop a mechanistic understanding of gas phase reactions and structure/property relationships of the catalyst.
- Investigate the effect of plasma parameters such as deposited power, temperature and residence time on concentration of particulate in exhaust.

Introduction

Previous work on this program showed that plasma-catalyst systems can reduce NO_X emissions in simulated diesel exhaust, however, improvements in the efficiency and design of the plasma reactor systems as well as in the efficiency of the catalysts is necessary for vehicle applications. Research in FY99 was aimed at improving catalyst and reactor efficiency as well as at understanding effects of real diesel exhausts on NO_X reduction activity. Research in FY99 focused on three general areas: catalyst development, reaction mechanism identification, and prototype reactor development.

Results

Systematic studies of structure property relationships in catalysts resulted in the discovery of a new catalyst with improved activity over a range of temperatures. As shown in Figure 1, the new catalyst (designated catalyst B) maintains 67% NO_X reduction activity up to 350°C, a 27%

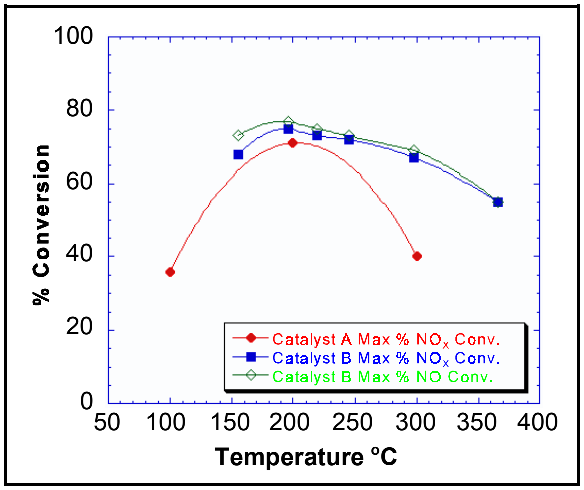

Figure 1. NO_X reduction as a function of temperature for new (B) and old (A) catalysts

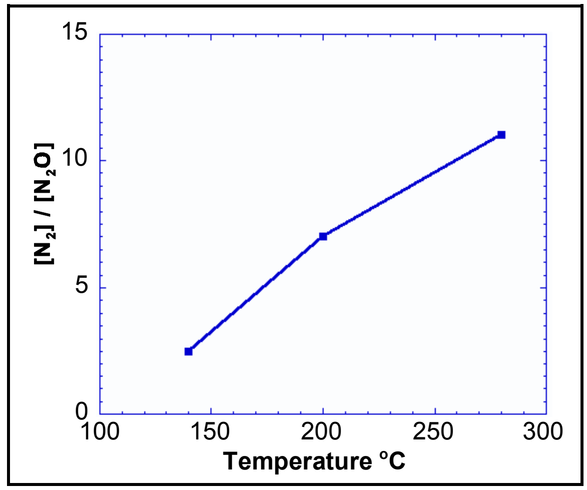

Figure 3. Ratio of nitrogen to N_2O formation as a function of temperature for new catalyst

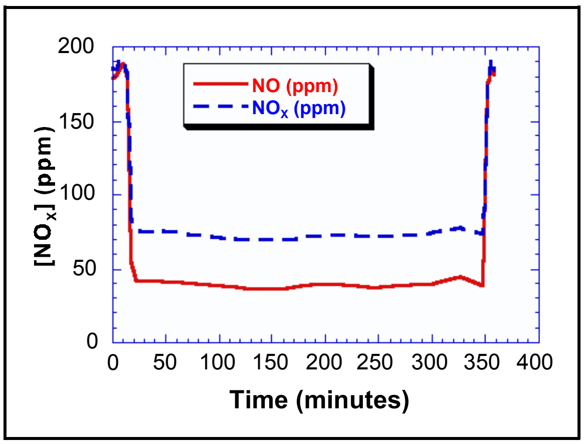

Figure 2. Data from 6 hours of a 30-hour test of NO_X reduction activity of Catalyst B as a function of time in simulated exhaust containing 50 ppm SO_2

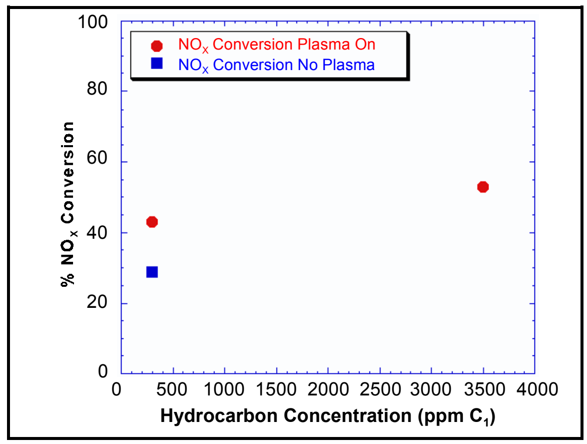

Figure 4. NO_X conversion using plasma assisted catalysis on diesel generator exhaust with propylene injection. 53% reduction is observed with an equivalent fuel penalty of 6%

improvement over a catalyst developed last fiscal year (designated catalyst A).

Because sulfur dioxide in the exhaust has been shown to rapidly poison many lean NO_X catalysts, the NO_X reduction activity of the new catalyst was tested in simulated exhaust with high sulfur dioxide content. As illustrated in Figure 2, this catalyst showed no degradation in activity after 30 hours of exposure to simulated diesel exhaust mix containing 50 ppm of SO_2.

A complete nitrogen balance was obtained for catalyst B. As shown in Figure 3, some N_2O is formed; however, N_2 is favored at all temperatures.

Bench-scale plasma-assisted catalyst reactor systems were scaled-up for slip-stream and full scale testing on exhaust from a diesel generator and from a VW TDI engine at ORNL. While more work is required to integrate a plasma device into a vehicle, this stand-alone device is the first device designed to treat the full exhaust stream from a diesel fueled vehicle. Full scale testing at two predetermined test points is ongoing. Up to 53% NO_X reduction at an equivalent fuel penalty of 6% was observed on a slip-stream of diesel generator exhaust as shown in Figure 4. Some improvement could be observed when hydrocarbon concentrations were increased by injecting propylene into the exhaust.

Conclusions

Plasma assisted catalyst systems can routinely reduce 70% NO_X on simulated diesel exhaust, however, NO_X reduction drops to 43-53% on real diesel exhaust from a diesel generator. In the last year a new catalyst with increased activity at high temperature and resistance to sulfur poisoning has been discovered. In addition a full scale plasma-catalyst system has been designed, fabricated and delivered for testing on a VW TDI engine at ORNL.

Publications

M.L. Balmer, R.G. Tonkyn, I. Yoon, S. Barlow, and J. Hoard, "Surface Mediation of NO_X Reduction/Oxidation in a Plasma," accepted Mat. Res. Soc. Proc., Fall Meeting Nov. 30-Dec. 4 1998.

J. Hoard, T.J. Wallington, J.C. Ball, M.H. Hurley, K.W. Wodzisz, and M.L. Balmer, "Role of Methyl Nitrate in Plasma Exhaust Treatment," accepted Environmental Science and Technology, October, April 1999.

M.L. Balmer, R. Tonkyn, A.Y. Kim, J. Hoard, I. S. Yoon, D. Jimenez, T. Orlando, and S. Barlow, "Nitrogen Measurement from NO_X Reduction for a Plasma-Catalyst System in Simulated Diesel Exhaust," Proceedings of the 1998 Diesel Engine Emissions Reduction Workshop in Castine, Maine, July, 1998.

J. Hoard and M.L. Balmer, "Plasma Catalysis on Simulated Diesel Exhaust," submitted to Applied Catalysis B: Environmental and Emerging Technology Reports, October, 1998.

J. Hoard and M.L. Balmer, "Plasma-Catalysis for Diesel NO_X Remediation," submitted to the Journal of Advanced Oxidation Technologies, October, 1998.

Patents

5,914,015 "Method and apparatus for exhaust gas..." issued 22 Jun 99, Stephan E. Barlow, Russel G. Tonkyn and Thomas M. Orlando.

5,746,984 "Exhaust System with Emissions Storage Device and Plasma Reactor", J.W. Hoard

E-1669, "Catalytic Plasma Reduction of NOx, " M. Lou Balmer, Russ Tonkyn, Andy Kim, Steve Barlow, and Thom Orlando. Filed September, 28 1998.

Presentations

M.L. Balmer, "Diesel NO_X Reduction Activity of Select Materials When Combined with a Plasma," Diesel Engine Emissions Reduction Workshop, Castine, ME, July 5-9, 1999.

J. Hoard, "Plasma Catalysis for Lean NO_X," Diesel Engine Emissions Reduction Workshop, Castine, ME, July 5-9, 1999.

M.L. Balmer, "New Technologies for the Reduction of Automobile Exhaust Emissions," DOE CIDI R&D Review, Argonne National Lab., June, 21-22, 1999.

M.L. Balmer "Surface Mediation of Nox Reduction/Oxidation in a Plasma," Materials Research Soc. Fall Meeting, Boston, MA, November 30-December 4, 1998.

M.L. Balmer "Diesel NO_X Reduction on Surfaces in Plasma," 1998 SAE International Fall Fuels and Lubricants Meeting and Exposition, San Francisco, California, October 19-22, 1998.

List of Acronyms

CIDI Compression Ignition Direct Injection
CRADA Cooperative Research & Development Agreement
LEP Low Emissions Technologies Research & Development Partnership
ORNL Oak Ridge National Laboratory
TDI Turbocharged Direct Injection

IV.C. Reduction of NO_X Emissions for Lean-Burn Engine Technology

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Objectives

- Develop new catalyst technology to enable CIDI engines to meet EPA Tier 2 emission standards.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 1A, Barriers A,B

Approach

- Development of new catalyst materials for reducing NO_X emissions in lean-burn exhaust environments, including:
  - Hydrous metal oxide-supported catalysts
  - Zeolite-supported catalysts
- Evaluation of new catalyst materials in both bulk powder and monolith forms, including short term durability testing, and testing of prototype catalytic converters on diesel engine dynamometers.
- Scale-up of synthesis and processing of promising catalyst formulations to enable fabrication of prototype catalytic converters for engine dynamometer testing.
- Technology transfer of most promising catalyst formulations and processes to LEP CRADA partners.

Accomplishments

- The Sandia portion of this project was the winner of the 1999 National Laboratory CIDI R&D Award in recognition of outstanding achievement in research and development of lean NO_X catalysts for CIDI emission control.
- Discovery by Los Alamos of a new family of zeolite-supported non-precious metal catalysts that have both high NOx reduction activity and a broad range of appreciable NO_X conversion.
- Optimized preparation procedures to enable reproducible catalyst fabrication using two different zeolite supports.
- Two U.S. patents have recently been issued on Sandia-developed materials. A U.S. patent application is being prepared for the new Los Alamos catalyst materials, covering formulations, preparation procedures, and potential catalytic applications.
- A new dopant for Pt/HTO:Si catalysts has been identified to improve catalyst performance by lowering the light-off temperature and widening the temperature window of appreciable NO_X reduction. A continuation-in-part patent application has been filed to cover these new catalyst formulations.
- A process was defined to enable transfer of the Sandia HMO-based catalyst material synthesis procedures to the LEP participants and their designated catalyst suppliers.
- The first phase of the technology transfer activities has been completed. Significant benefits achieved by the technology transfer process were that these efforts added to the catalytic materials knowledge base and provided new insight, as well as providing a potential payoff with other catalyst materials and/or applications.
- The engine dynamometer test bed was transitioned from a larger 7.3 L Navistar diesel engine to a 1.9 L Volkswagen TDI diesel engine, which should be more representative of the engine-out emissions profile of the prototype PNGV vehicles.
- Several catalysts were evaluated on the new 1.9 L VW TDI diesel engine dynamometer.
- It was determined that significantly lower NO_X reduction activity was obtained for all catalysts tested on the VW TDI engine relative to the Navistar engine. This was possibly related to higher particulate matter emissions from the VW engine detrimentally affecting catalyst performance.

Future Directions

- Completion of the experimental matrix evaluating fundamental aspects, processing and durability of the new Los Alamos catalyst materials.
- File U.S. patent application related to new Los Alamos catalyst materials, covering the required preparation procedures and potential catalytic applications.
- Technology transfer to the LEP of the new Los Alamos catalyst formulations and processing procedures to facilitate the fabrication of full-scale converter monoliths for engine dynamometer testing. Complete second phase of technology transfer involving Sandia catalyst technology.
- Determine the possible effects of particulate matter on the NO_X reduction performance of lean NO_X catalysts.
- Address new and potentially more efficient NO_X reduction options for lean-burn exhaust aftertreatment, including:
    - Selective catalytic reduction of NO_X by urea (or NH_3);
    - Mixed catalyst and support studies designed to optimize lean-burn NO_X catalysts;
    - Fundamental studies related to determining the mechanism of the selective catalytic reduction of NO_X over different catalyst materials with various reductants.

Introduction

This multi-partner effort has been continued under OAAT sponsorship and involves separate CRADAs between three national laboratories (Los Alamos National Laboratory [LANL], Oak Ridge National Laboratory [ORNL], and Sandia National Laboratories [SNL]) and the Low Emission Technologies Research and Development Partnership (LEP, composed of DaimlerChrysler Corporation, Ford Motor Company, and General Motors Corporation). The project addresses reduction of CIDI engine NO_X emissions using exhaust aftertreatment—identified as one of the key enabling technologies for CIDI engine success. The overall CRADA efforts are focused on the development and evaluation of new catalyst materials for reducing NO_X emissions, specifically targeting the selection of appropriate catalyst materials to meet the exhaust aftertreatment needs of the PNGV vehicles.

With the evolution of the new EPA Tier 2 emission standards scheduled to be in place starting in 2004, the program has now begun general redirection efforts to address new and potentially more efficient NO_X reduction options for lean-burn exhaust aftertreatment. New possible efforts involve the investigation of catalysts for use in the selective catalytic reduction of NO_X by urea (or NH_3), as well as more fundamental studies related to determining the mechanism of the selective catalytic reduction of NO_X over different catalyst materials with various reductants, and mixed catalyst and support studies designed to optimize lean-burn NO_X catalysts.

LEP Partner Work-In-Kind Efforts

Considerable automaker effort over the last year has identified a laboratory gas mix that more effectively simulates the performance of a modern, small, high-speed CIDI engine running on conventional diesel fuel in terms of engine-out emissions. This new gas mix formulation will enable all of the national laboratories to better simulate diesel engine dynamometer performance using conventional flow reactor units. A comparison of the new and old (simulated lean-burn gasoline exhaust) mix formulations is shown below in Table 1.

Table 1. Laboratory gas mix formulations representative of lean-burn gasoline (old mix) and small displacement CIDI (new mix) engine exhaust compositions

Species	New Mix	Old Mix
NO_X	75	250
Total HC (ppm as C1)	600	2100
n-C_8H_{18} (ppm as C1)	450	525 (C_3H_8)
C_3H_6 (ppm as C1)	150	1675
Total HC (ppm as C1): NO_X	8:1	8.4:1
CO (ppm)	600	400
H_2 (ppm)	200	133
CO_2 (%)	5.0	7.0
O_2 (%)	12	8
H_2O (%)	5.0	8.0
SO_2 (%)	1.5	15
Space Velocity (cc/cc*h^{-1})	25,000	50,000

Los Alamos National Laboratory (LANL) Efforts

LANL's work has focused on the development of new and stable zeolite-based catalysts for NO_X reduction in lean-burn exhaust environments. This work is complementary to the SNL work since these non-precious metal catalysts are active for NO_X reduction at higher temperatures than those of the Pt-based catalysts under investigation at SNL.

Screening experiments initiated in FY98 involving microporous support materials resulted in the discovery of a new family of zeolite-supported non-precious metal catalysts that have both high NO_X reduction activity and a broad range of appreciable NO_X conversion. After discovery of this new family of materials in early FY99, experiments focused on the optimization of preparation procedures to enable reproducible catalyst fabrication. These experiments involved the choice of zeolite support (Support 1 or Support 2), support pretreatment procedure (Treatments A-I), choice of the active metal catalyst (Metal 1 or Metal 2), and the method of exchange (Exchange Method 1 or Exchange Method 2) used to prepare the zeolite-supported non-precious metal catalyst. A U.S. patent application is being prepared for these new catalyst materials, covering formulations, preparation procedures, and potential catalytic applications.

An example of the importance of catalyst preparation process variables is illustrated in Figure 1. This figure shows the effect of similar support pretreatment procedures (Treatments A-D) on two different zeolite supports used to fabricate catalysts containing Metal 1 added via Exchange Method 1. These results are very interesting in that different pretreatment procedures are observed to optimize catalyst performance for Support 1 (Treatment A) and Support 2 (Treatment C). This obviously stresses the importance of understanding the role of the interaction of catalyst preparation variables on NO_X reduction performance.

Plans for future work at LANL include the following: completion of the experimental matrix evaluating the various catalyst preparation variables, understanding the adsorption vs. catalysis functionality of these new materials, and short term durability testing of these new catalysts at high temperature in environments containing steam and/or SO_2. Technology transfer to the LEP of these new catalyst formulations and processing procedures is planned. This will enable the

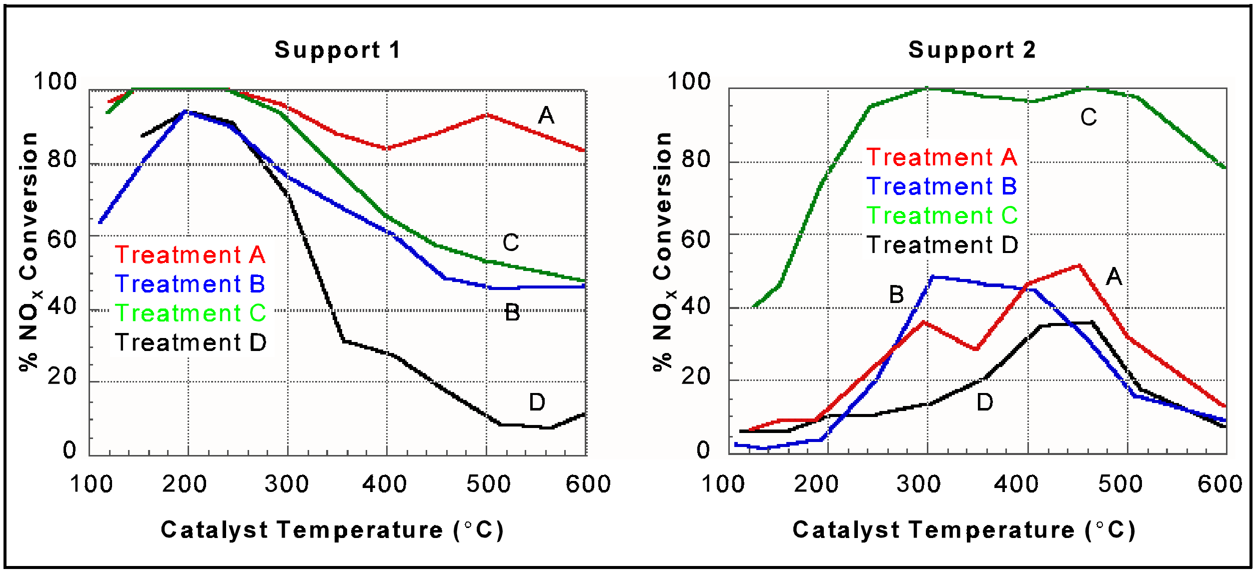

Figure 1. Results showing the effect of support pretreatment on NO_X conversion activity for catalysts synthesized using two different zeolite supports

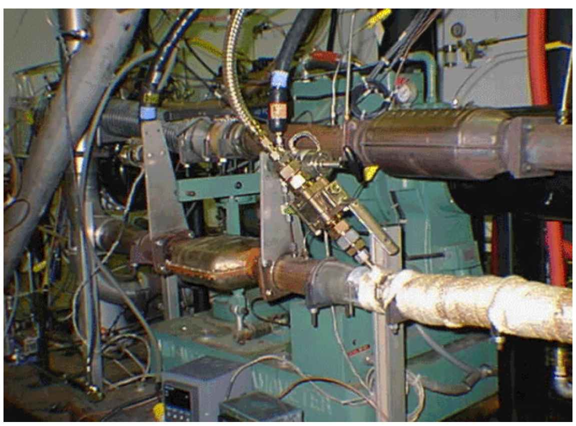

Figure 2. Exhaust and catalytic converter layout used in conjunction with the VW 1.9 L TDI diesel engine test bed at Oak Ridge National Laboratory

fabrication of full-scale converter monoliths for engine dynamometer testing.

Oak Ridge National Laboratory (ORNL) Efforts

ORNL's continuing role in this project has been to provide characterization of catalyst performance, both in bench scale reactor testing and in an engine laboratory, in addition to microstructure characterization of catalysts using electron microscopy.

At the suggestion of the industry partner team, the test bed was transitioned from the larger 7.3 L Navistar diesel engine, which had served for over four years in the project, to the 1.9 L Volkswagen (VW) TDI diesel engine. The engine-out emissions profile of this new engine is more representative of the conditions that will exist in the prototype PNGV vehicles. Therefore, it was important to more accurately represent the conditions that these catalysts will experience, especially as the catalysts become more refined to the particular application. Figure 2 shows the exhaust/converter system for the VW 1.9 L TDI diesel engine test bed in the ORNL engine laboratory.

Over the last year, several catalysts were evaluated on the new 1.9 L VW TDI diesel engine dynamometer, with results compared to the previous performance of the same catalyst on the 7.3 L Navistar diesel engine dynamometer. The lean-NO_X catalytic converter used with the new Volkswagen CIDI engine was benchmarked as a reference point (designated Vendor #1 catalyst). Various

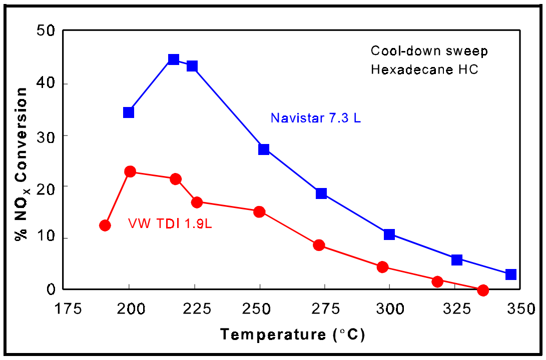

Figure 3. Vendor #1 catalyst performance for NO_X reduction. Both catalysts were evaluated at a space velocity of 50,000 hr^{-1} and an HC (as C1):NO_X ratio of 8:1

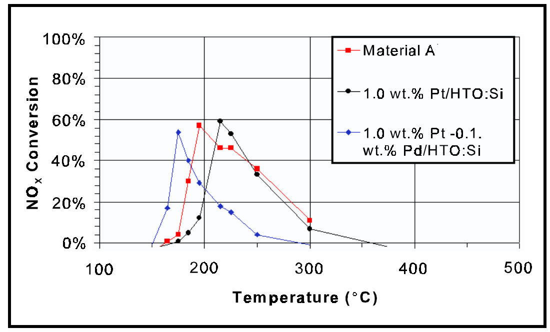

Figure 4. Comparison of NO_X reduction results for various Pt-based/HTO:Si catalysts using the old mix gas formulation. Material A represents a new dopant added to the baseline Pt/HTO:Si material.

hydrocarbon reductants were used during the evaluation, including hexadecane and conventional diesel fuel.

Results for the Vendor #1 catalyst tested behind both the Navistar and VW TDI engines using a hexadecane reductant are compared in Figure 3. Significantly lower NO_X reduction activity was obtained for all catalysts on the VW TDI engine relative to the Navistar engine. However, the difference in catalyst performance between the two engine exhaust environments could not be attributed to the lower engine-out NO_X obtained with the VW TDI engine. In conjunction with the lower NO_X conversion, significantly lower hydrocarbon conversion was also obtained in the case of the VW TDI engine. Significant sooting of the catalyst monolith frontal surface and inside channels after VW TDI engine dynamometer testing indicated that particulate matter (PM) emissions from this engine might be affecting catalyst performance. Higher PM emissions might be expected for this engine since the exhaust gas recirculation (EGR) mode was active for all test conditions. Further experiments are in progress to better understand these results and to determine the possible effects of PM on the NO_X reduction performance of lean-NO_X catalysts.

Microstructural evaluation continues to be an integral part of the CRADA, both for catalyst development and characterization before and after engine laboratory testing. Significant work was performed over the last year on the new SNL Material A (described in the next section), both in bulk powder and monolith core form.

Sandia National Laboratories (SNL) Efforts

Significant progress was made related to new catalyst development efforts. A new dopant for silica-doped hydrous titanium oxide-supported Pt (Pt/HTO:Si) catalysts has been identified to improve catalyst performance by lowering the light-off temperature and widening the temperature window of appreciable NO_X reduction (see Material A in Figure 4). Optimization studies on both bulk and monolith core catalysts have been completed. Significant characterization efforts are in progress (including the microstructural characterization efforts described above) to understand the role of the dopant in altering catalyst performance. This material has been fabricated in monolith core form using both the direct coating and powder slurry methods.

Significant effort over the past year has been devoted to technology transfer. The first phase of the transfer of the SNL HMO-supported catalyst technology to the LEP and designated catalyst suppliers has now been completed. A process was defined to enable transfer of the HMO-based catalyst material synthesis procedures to the participants, and signed nondisclosure agreements are in place with four major catalyst suppliers. This phase of the technology transfer activities involved separate site visits (to SNL) with the LEP members and their designated catalyst suppliers, the development of the necessary process documentation to support technology transfer efforts, and feedback from the separate OEM/supplier teams regarding their efforts in duplicating the SNL technology. The various catalyst suppliers have completed their initial evaluation of the SNL HMO-supported catalyst technology and made decisions regarding their future efforts in this area. In general, it can be summarized that a significant breakthrough regarding lean NO_X catalysis was not achieved with the SNL materials and process, although this result is more a function of the extremely aggressive EPA Tier 2 emissions standards than the specific attributes of Pt-based/HTO:Si catalysts. Significant benefits achieved by the technology transfer process were that these efforts added to the catalytic materials knowledge base and provided new insight, as well as providing a potential payoff with other catalyst materials and/or applications. Technology transfer activities are continuing with a second phase that involves special additives to Pt/HTO:Si catalysts (Material A) which both lower light-off temperature and broaden the temperature window of appreciable NO_X conversion. It is hoped that these additives will be beneficial in a generic sense so they can be applied to a variety of supported Pt catalysts.

Two U.S. patents have recently issued on SNL-developed materials. One patent (U.S. 5,795,553 issued on August 18, 1998) was related to a nitrogen oxide adsorbing material, while the other (U.S. Patent 5,830,421 issued on November 3, 1998) was related to nitrogen oxide reduction catalysts. A continuation-in-part patent application has also been filed to cover the new catalyst formulations with a lower light-off temperature and a wider temperature window of appreciable NO_X conversion. This project was the winner of the 1999 National Laboratory CIDI R&D Award in recognition of outstanding achievement in research and development of lean NO_X catalysts for CIDI emission control.

Presentations/Publications/Patents

S. E. Lott, T. J. Gardner, L. I. McLaughlin, C. A. Matlock, and J. B. Oelfke, U.S. Patent No. 5,795,553, Nitrogen Oxide Adsorbing Material, August, 18, 1998.

N. C. Clark, J. A. Rau, K. C. Ott, and M. T. Paffett, "The Role of Hydrocarbon Reductant in Metal Entrained Zeolite and ALPO DeNox Catalysis", presented at the American Chemical Society National Meeting, Boston, MA, August 23, 1998.

T. J. Gardner, S. E. Lott, S. J. Lockwood, and L. I. McLaughlin, Serial No. 09/185,149, Material and System for Catalytic Reduction of Nitrogen Oxide in the Exhaust Stream of a Combustion Process, November 2, 1998.

T. J. Gardner, S. E. Lott, S. J. Lockwood, and L. I. McLaughlin, U. S. Patent No. 5,830,421, Material and System for Catalytic Reduction of Nitrogen Oxide in an Exhaust Stream of a Combustion Process, November 3, 1998.

T. J. Gardner, L. I. McLaughlin, R. S. Sandoval, J. G. Reynolds, K. C. Ott, M. T. Paffett, N. C. Clark, J. A. Rau, R. G. McGill, N. Domingo, J. Storey, and K. L. More, "Reduction of NO_X Emissions for Lean-Burn Engine Technology: A Cooperative Research Effort Between the National Laboratories and the U.S. Auto Industry," Poster Presented at the DOE Laboratory Catalysis Research Symposium, Albuquerque, NM, February 24, 1999.

L. I. McLaughlin, T. J. Gardner, and R. S. Sandoval, "Hydrous Metal Oxide-Supported Catalysts for Automotive Exhaust Treatment Applications," Presented at the 13th Annual Meeting of the Western States Catalysis Club, Albuquerque, NM, February 26, 1999.

M. T. Paffett, N. C. Clark, J. A. Rau, and K. C. Ott, "The Role of Hydrocarbon Reductant in Metal Entrained Zeolite and ALPO DeNOx Catalysts," Presented at the 13th Annual Meeting of the Western States Catalysis Club, Albuquerque, NM, February 26, 1999.

T. J. Gardner, L. I. McLaughlin, and R. S. Sandoval, "Hydrous Metal Oxide-Supported Catalysts for Automotive Exhaust Treatment Applications," Poster Presented at the Sixteenth North American Meeting of the Catalysis Society, Boston, MA, May 31, 1999.

K. C. Ott, N. C. Clark, and M. T. Paffett, "Reactivity and Short Term Stability of Zeolite-Based Lean NO_X Catalysts with Sulfur Dioxide and Water Under Simulated Automobile Exhaust," Presented at the Sixteenth North American Meeting of the Catalysis Society, Boston, MA, June 1, 1999.

Acronyms

CIDI	Compression Ignition Direct Injection
CRADA	Cooperative Research and Development Agreement
EGR	Exhaust Gas Recirculation
EPA	Environmental Protection Agency
HC	Hydrocarbon
HMO	Hydrous Metal Oxide
HTO:Si	Silica-Doped Hydrous Titanium Oxide
LANL	Los Alamos National Laboratory
LEP	Low Emissions Technologies Research and Development Partnership
NO_X	Nitrogen Oxides (NO and NO_2)
OEM	Original Equipment Manufacturer
ORNL	Oak Ridge National Laboratory
PM	Particulate Matter
PNGV	Partnership for a New Generation of Vehicles
ppm	parts per million (volume basis)
R&D	Research and Development
SNL	Sandia National Laboratories
TDI	Turbo Direct Injection
VW	Volkswagen

IV.D. Catalyst Applications of High-Field Microwave Energy

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Objectives

- Reduce NO_X emissions by coupling high-field microwave energy to catalyst applications

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 1A, Barriers A,B

Approach

- Improve NO_X conversion by applying high-field microwave energy directly to Pd and Pt based-catalysts.
- Provide a proof-of-principle demonstration of nonthermal (rotational) excitation by evaluating catalyst performance as a function of the peak and average applied power (J).
- Evaluate the conversion of NO_X in a plasma generated via high-field microwave application.

Accomplishments

- A microwave catalyst exhaust measurement system (MW generator, bench-flow reactor, analyzers) was built and tested.
- Preliminary testing showed a rapid conversion of CO to CO_2 with the application of high- field microwave energy.
- Measured the formation of NO, NO_2 and N_2O as a function of energy density for a microwave-generated plasma (feedgas was 12 %O_2 in N_2).

Future Directions

- Evaluate the influence of hydrocarbon and water of the formation of NO_X during plasma generation.
- Provide a proof-of-concept demonstration of the nonthermal effect.
- Perform tests to measure the conversion chemistry and efficiencies of CO and NO_X for catalysts exposed to high-field microwave excitation.

Introduction

The interactions between catalyst and high-field microwave energy are being investigated as a means of increasing the conversion of NO_X in material catalysts. Microwave heating provides a feasible means of rapid heating on material surfaces. When used in the high-field mode, microwave energy may cause rotational excitation on the catalyst surface, thereby absorbing even higher energy levels which may enhance surface reactions associated with catalysis.

A microwave catalyst testing apparatus consisting of a microwave source and wave guide, programmable bench-flow reactor, and FTIR and other analyzers, which are shown

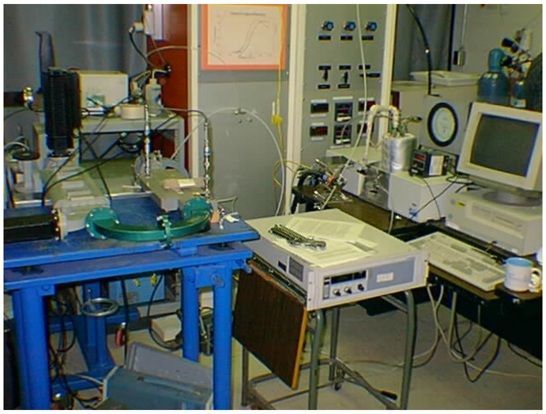

Figure 1. Assembled microwave catalyst system including bench-flow reactor, FTIR spectrometer, and CO gas analyzer

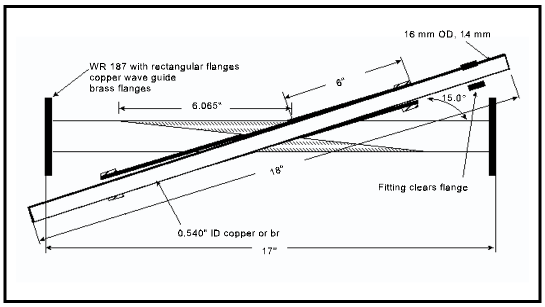

Figure 2. Schematic diagram of microwave test chamber

together in Figure 1. The test chamber is incorporated into the waveguide as shown in Figure 2.

Microwave Plasma Generation and Production of Unwanted Species

The formation of NO, NO_2, and N_2O in a microwave generated plasma was performed under the conditions outlined in Table 1 and measured using an FTIR spectrometer. The peak power and pulse length were held constant.

Energy density was controlled by varying the period freqency. The feed gas consisted of 12 % O_2 in N_2 and was maintained at a flow rate of 1500 cc/minute. As shown in Figures 2 through 4, the amount of NO, NO_2, and N_2O decreased to near zero levels with decreasing

Table 1. Energy Density Test Conditions

Test Case	Peak Power (kW)	Pulse Width (μs)
1	80	0.5
2	80	0.5
3	103	1.7
4	103	1.7

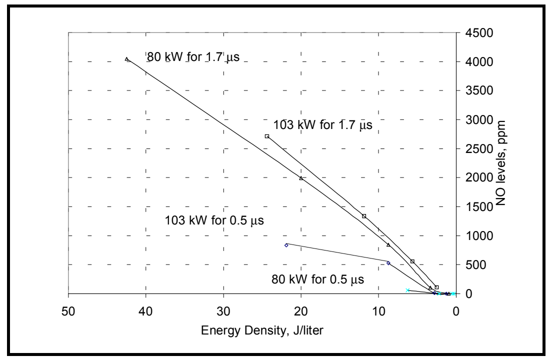

Figure 3. NO formation associated with energy density for a high-field microwave generated plasma

energy density. Of interest, the formation of NO (Figure 2) shows a dependency on the peak power (NO increases with increasing peak power), whereas the formation of NO_2 and N_2O do not. Measurable levels of N_2O_5 were observed for the 80kW (0.5μs) test case but not for the other test cases.

A simulated diesel emission was tested under the low energy density plasma conditions. No conversion of NO to NO_2 was observed.

Conclusions

The formation of NO, NO_2, and N_2O in plasmas generated by high-field microwave energy could be controlled to near zero levels using very low energy densities. Yet, the conversion characteristics of NO do not indicate that high-field plasmas offer an

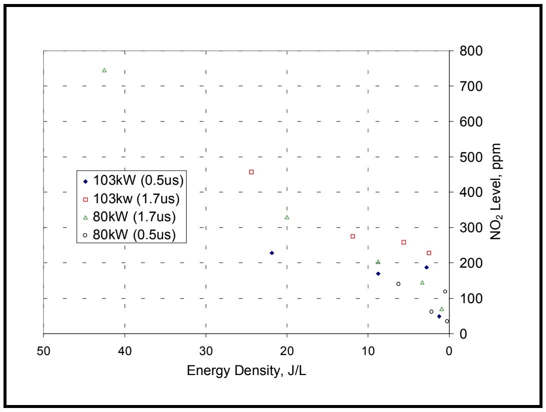

Figure 4. NO_2 formation associated with energy density for a high-field microwave generated plasma

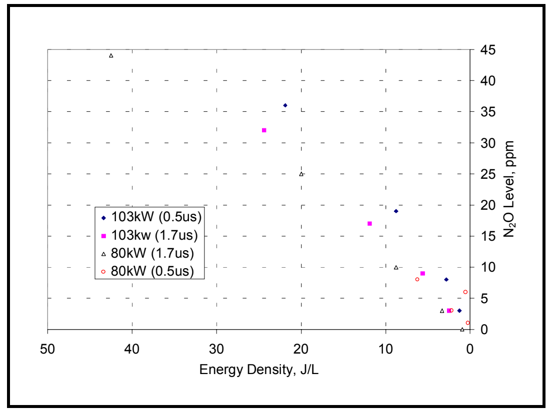

Figure 5. N_2O formation associated with energy density for a high-field microwave generated plasma

advantage over corona-discharge plasmas. The direct application of high-field microwave energy to the catalyst should produce nonthermal excitation which should improve the conversion efficiency of NO_X in diesel emissions.

Presentations

Michael D. Kass, John M. Storey, Greg Hanson, and John Whealton, "Effect of Microwave Fields on Catalysts: Direct and Indirect Effects, Diesel Combustion & Aftertreatment R&D Laboratory Merit Review and Peer Evaluation," Argonne National Laboratory, Chicago, Illinois, June 21-23, 1999.

Michael D. Kass, John M. Storey, Greg Hanson, and John Whealton, "Applications of Microwave Fields to Catalyst Research," Diesel Engine Emission Reduction Workshop 99, Castine, Maine, July 5-8, 1999

IV.E. NO_X Adsorber Catalyst Project

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Objectives

- Develop and implement full-scale prototype of sulfur tolerant NO_X adsorber for demonstration on a light-duty CIDI engine.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 1A, Barriers A,B

Approach

- Develop and test a NO_X/SO_X adsorber system that can be upscaled to a prototype.
- Develop and test a prototype combustor to provide a source of hydrogen gas used for catalyst regeneration.
- Test and model the performance of the adsorber system with the combustor on an actual operating engine.

Accomplishments

- A prototype combustor has been fabricated.
- The adsorber system was demonstrated to reduce NO_X emissions to near zero levels for high ppm levels of NO_X, high space velocities, and a wide temperature range.
- The adsorber system was demonstrated to be efficiently regenerated by hydrogen gas.

Future Directions

- Implement the combustor on a diesel engine and measure/model the formation of hydrogen and other species.
- Test sulfur resistant catalyst in combination with a regeneration process that uses diesel fuel.

Introduction

NO_X adsorber catalysts work by storing NO_X over a period of time. Once the catalyst is saturated it is then regenerated by hydrogen and/or hydrocarbons. The prototype combustor (shown in Figure 1) was fabricated to generate hydrocarbon and hydrogen species from diesel fuel. Additionally, the sorption characteristics of the SO_2 and NO_X adsorbers were determined using bench-scale experiments. These experiments used the simulated diesel emission, the components of which are listed in Table 1.

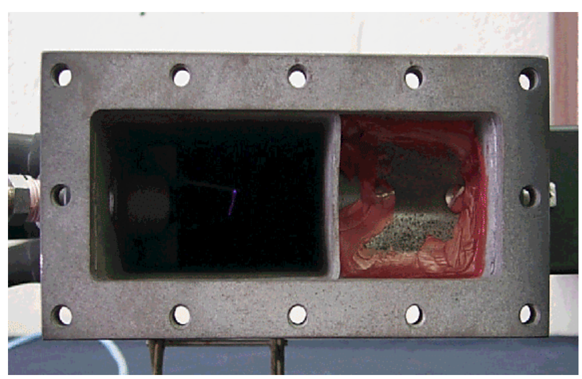

Figure 1. Prototype diesel fuel combustor developed to provide regeneration gas mixture for adsorber. Note arcing in the center of the dark rectangle.

Table 1. Simulated Diesel Exhaust

Component	Level
NO_X	300 ppm
CO	600 ppm
O_2	45 ppm
CO_2	12%
H_2O	5%
SO_2	5%
N_2	balance

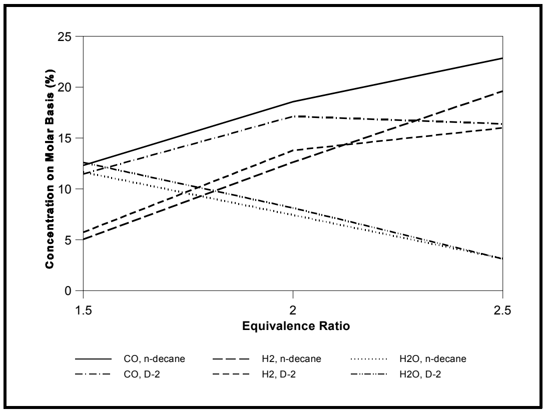

Figure 2. Calculated composition versus equivalence ratio for combustor products

Combustor Development

The combustor shown in Figure 1 was tested and modeled for lean burn conditions. The product composition as a function of equivalence ratio for this combustor was calculated and is shown in Figure 2.

SO_2 and NO_X Adsorber Bench-Scale Testing

Bench-scale testing was performed to evaluate adsorber performance as a function of the NO_X level, space velocity, and temperature. Regeneration of the adsorbers was performed using a mixture containing 4% H_2 and 1% H_2O in N_2. The space velocities during regeneration were 8,000/h and 16,000/h. The results of the NO_X level, space velocity, and

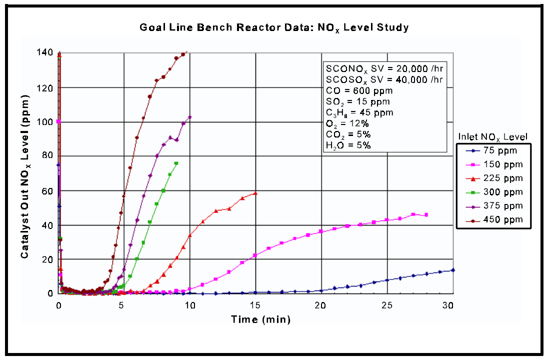

Figure 3. Adsorber performance versus the level of NO_X

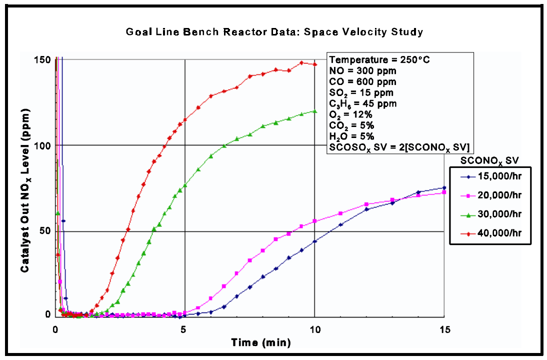

Figure 4. Adsorber performance versus space velocity

temperature are shown in Figures 3, 4, and 5 respectively. The adsorber system was tested using NO_X levels up to 450 ppm while controlling the space velocity at 20,000/h and 40,000/h. The results, which are shown in Figure 3, show that the NO_X levels were reduced to near zero levels even at 450 ppm NO_X. The space velocity test results are shown in Figure 4. In this study, the combustor reduced 300 ppm of NO_X to near zero values even at a space velocities up to 40,000/h. As shown in Figure 5 the adsorber was effective over a wide temperature range for the same 300 ppm level of NO_X.

Engine Test Results

NO_X conversion was measured on a light-duty CIDI engine as a function of exhaust temperature. As shown in Figure 6 the conversion efficiencies were exceptionally high

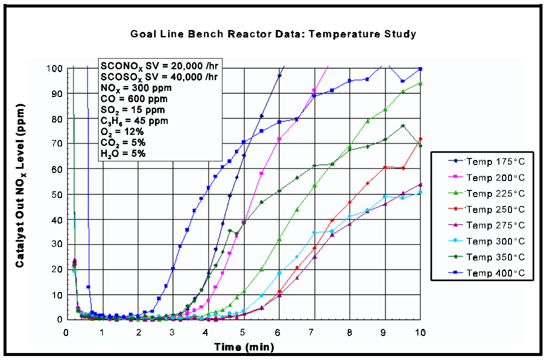

Figure 5. Adsorber performance as a function of temperature

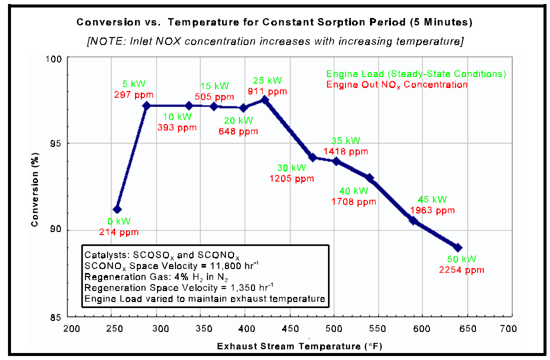

Figure 6. Engine Test Results

for exhaust temperatures between 275 and 500°C.

Conclusions

In conclusion, the test results show NO_X was reduced to near zero levels by the adsorber system even at high space velocities, high NO_X levels. The adsorber system performed well over a wide temperature range indicating that this technology is an effective means of reducing NO_X emissions from diesel emissions.

Presentations

Michael D. Kass, John M. Storey, Greg Hanson, and John Whealton, "NO_X Adsorber Catalyst Project," Diesel Combustion & Aftertreatment R&D Laboratory Merit Review and Peer Evaluation," Argonne National Laboratory, Chicago, Illinois, June 21-23, 1999.

List of Acronyms

CIDI	Compression Ignition Direct Injection
NO_X	Oxides of Nitrogen
SO_X	Oxides of Sulfur

IV.F. Catalyst Surface Diagnostics

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DOE Program Manager: Ken Howden, (202) 586-9811, fax: (202) 586-3631, e-mail: ken.howden@ee.doe.gov

Objectives

- Improve understanding of NO_X reduction mechanisms for application to development of advanced catalyst/reductant processes.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 1A, Barriers A,B

Approach

- Apply spectroscopic diagnostics to investigate the nature of surface species participating in catalytic reduction and the effectiveness of individual HCs on lean NO_X reduction.
- Develop a high-speed diagnostic for quantifying exhaust nitrogen species for application to resolving the nitrogen balance.

Accomplishments

- The signal-to-noise ratio of the Diffuse Reflectance mid-Infrared Fourier Transform (DRIFT) spectrometer was significantly improved by incorporating an MCT detector.
- The transient nature of surface-adsorbed species relevant to the catalytic process of interest was evaluated using DRIFT spectroscopy.
- The excitation/detection scheme of the Raman spectrometer was modified to mitigate broad and distinct interfering signals.
- A test cell for use with the Raman spectrometer and to mimic the flow at a channel wall of a catalyst brick was developed and tested.
- A field-applicable, mass-spectroscopy based Fast-NO_X instrument was developed and deployed to measure exhaust NO_X in an engine test cell.

Future Directions

- Apply diagnostics to identify relevant mechanisms via bench and engine experiments. Develop high speed, high sensitivity measurements for surface species in real systems using advanced spectroscopic techniques.

Catalyst Diagnostics

The current experiments are focused on obtaining diffuse reflectance (DRIFT) and Raman spectra of adsorbed molecules on catalyst surfaces. A schematic of the DRIFT instrument is shown in Figure 1 and features a barrel ellipse configuration with a highly sensitive mercury- cadmium-telluride (MCT) detector. The quality of the spectra have been consistently high with the new detector, and a variety of materials, provided by the industrial partner, are being evaluated. The Raman cell is shown in Figure 2 and features a heated portion with a quartz window for spectroscopic use. Materials and substrates can be analyzed through the window. A number of different

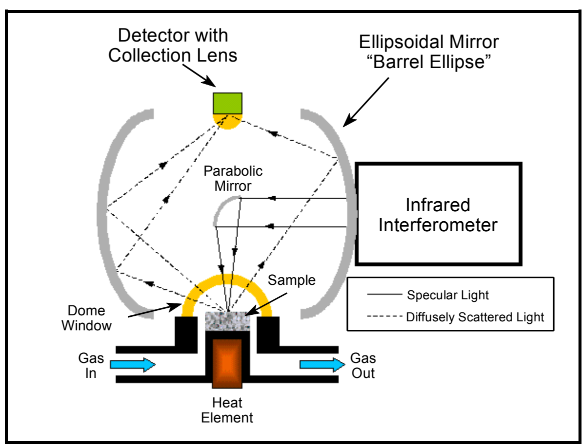

Figure 1. Schematic of the Diffuse Reflectance mid-Infrared Fourier Transform (DRIFT) Spectrometer

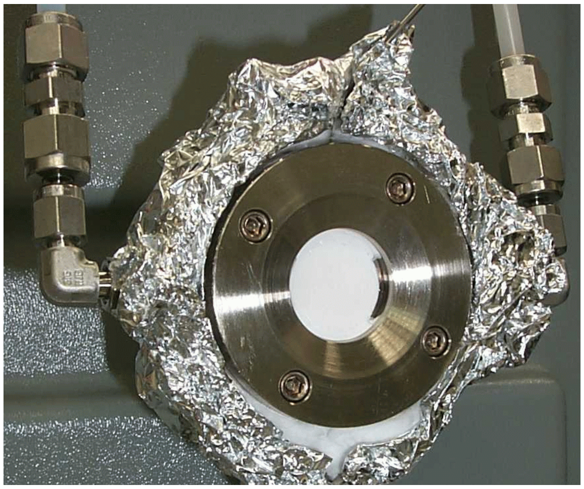

Figure 2. Raman cell to simulate channel-wall flow

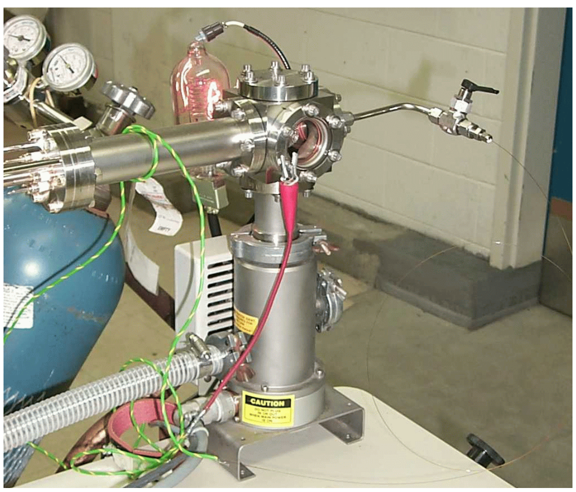

Figure 3. Mass-spectrometer Based Fast-NO_X Instrument

laser Raman configurations have been evaluated with the material often dictating the appropriate wavelength and detection scheme.

High-Speed NO_X Detection

The mass-spectroscopy based Fast-NO_X instrument is shown in Figure 3. A narrow bore capillary physical probe is used for minimally-invasive gas-stream sampling and delivery to the ionization chamber (apparent in the center of Figure 1). A turbo pump is situated below, and a quadrapole detector to the left of the ionization chamber.

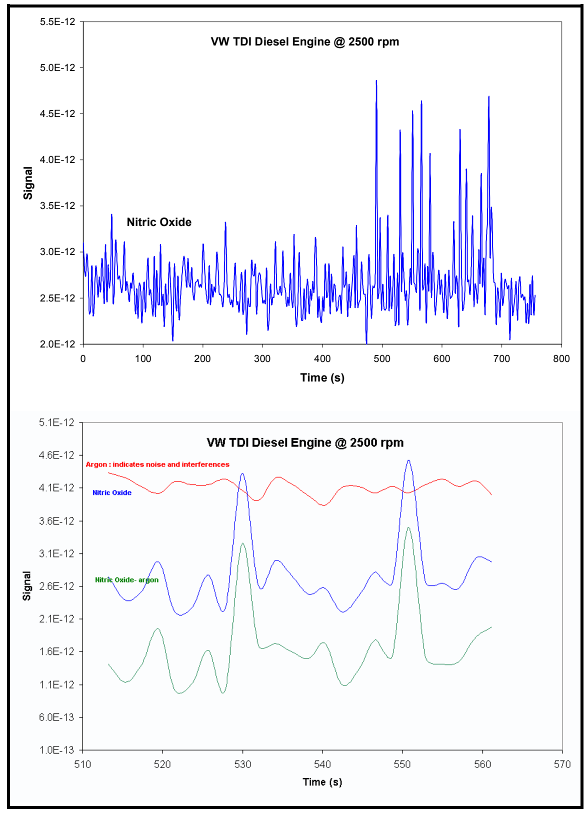

Figure 4. Trace of instrument response to transient engine operation. Detail plot shows comparison with argon signal. Subtraction of the argon signal shows NO freed of pressure effects.

We have tested the instrument using fuel injectors to supply discrete pulses of NO (1000 ppm) in zero gas, simulating dynamic exhaust concentration fluctuations over three orders of magnitude. These test indicated an ability to resolve temporal fluctuations on the order of 3 Hz; this is a significant improvement beyond that achievable with standard chemiluminescence measurements.

The Fast-NO_X instrument has been demonstrated using an actual exhaust matrix generated from a running Volkswagen TDI diesel engine. The exhaust sample was extracted downstream from a de-activated catalyst. Additional insights are apparent from Figure 4, which shows dynamic measurements of argon and nitric oxide from the engine test.

The argon measurements characterize both the system noise and anomalous signal contribution due to pressure fluctuations. The engine was run at a steady state, low load for the first 480 seconds of the test. At 480 seconds into the test, emissions fluctuations were induced by momentarily increasing the fueling rate at an approximately constant repetition rate. The fueling-rate spikes were terminated at 580 seconds and reinitiated at a higher repetition rate from 620 to 680 seconds. Nitric oxide fluctuations resulting from the fueling spikes are readily apparent above the measurement noise as characterized by the argon measurements. Moreover, the argon measurements indicate no anomalous signal contribution due to pressure fluctuations; such an interfering contribution to the nitric oxide measurements would be indicated by a synchronous spike in the argon signal level. In addition to the induced dynamics, the data appears to indicate real nitric oxide fluctuations in the steady-state operation regime. This data provides validation of the instrument's ability to make measurements in the harsh environment of a realistic exhaust stream.

Summary

The catalyst diagnostics have moved from a design/evaluation phase into application to partner- supplied materials. The Fast-NO_X instrument has been demonstrated on real diesel exhaust.

V. EXHAUST GAS PARTICULATE EMISSION CONTROL R&D

V.A. Exhaust Port Particulate Matter Measurements

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Objectives

- Develop a real-time, engine-out particulate matter (PM) diagnostic for size, number density and volume flux that can be easily transferred to industry

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 1C, Barriers A,B

Approach

- Simultaneous measurements of Laser Induced Incandescence and Rayleigh Scattering (LII-RS) are used to obtain the PM volume fraction, primary particle size, and volume-equivalent-sphere diameter.
- Off-the-shelf components are used to build a measurement system that can be easily duplicated by industry partners.

Accomplishments

- A 4-channel, 500 MHz bandwidth, 5 GS/s, PC-based data acquisition system is operational.
- A variety of fast, compact, off-the-shelf photodetectors have been evaluated for detecting the LII and RS signals.
- LII signals have been characterized in a propane diffusion flame.
- A first-generation cell for optical access to the exhaust port was designed, built, and tested.
- Preliminary LII measurements have been obtained in a spark-ignition engine during a cold start, snap-throttle test.

Future Directions

- Quantify the LII-RS measurements.
- Develop a diagnostic for instantaneous exhaust velocity to obtain volume flux.
- Apply the LII-RS technique to diesel exhaust.

Introduction

LII is a fairly well-established technique for the measurement of PM volume fraction; it has been applied to both stationary burner flames and diesel engine combustion. Light from a high-energy pulsed laser is used to quickly heat the PM to its vaporization point, resulting in gray-body radiation I_{LII} that is proportional to the PM volume fraction. Simultaneous measurement of Rayleigh scattered light I_{RS} from the particles can be used to obtain the volume-equivalent-sphere diameter d and the particle number density N, where

$$d \sim (I_{RS} / I_{LII})^{1/3} \quad \text{and} \quad N \sim (I_{LII})^2/I_{RS}$$

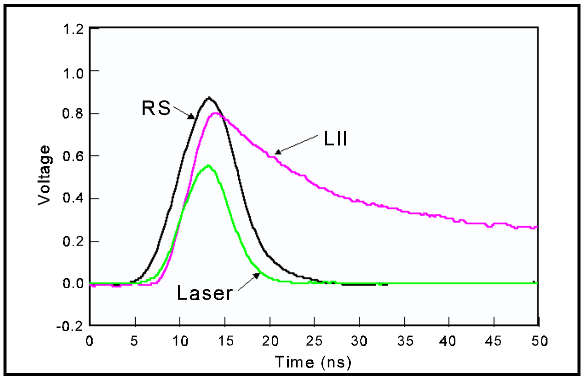

Figure 1. Typical LII-RS signals obtained in a propane diffusion flame for a single laser pulse

In addition, the decay rate of the LII intensity following the laser pulse can be used to determine the primary particle size, since it is proportional to the rate of heat loss from the particle. Typical signals obtained in a propane diffusion flame are presented in Figure 1.

PM Measurement System

It is important to note the very fast time scales of the signals shown in Figure 1. The measurements of interest are the areas under the laser and RS curves, and the maximum of the LII curve and its decay rate. The first three measurements can be made with a fast gated-integrator, and a fourth, delayed gated-integrator measurement of the LII curve can be used to approximate the decay rate. However, for considerably less cost one can buy a digital real-time oscilloscope that will record all of the data shown in Figure 1, permitting thorough post-processing of the signals. By using this approach, the majority of the development work goes into computer software, resulting in a near turn-key system that can be easily passed on to industry for their use.

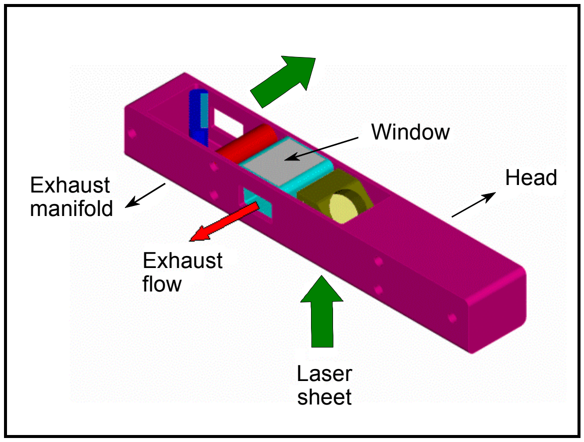

Figure 2. Exhaust-port optical cell

To obtain optical access to the exhaust flow close to the port, SNL designed the optical cell shown in Figure 2. It consists of a spacer that installs between the head and exhaust manifold. Our single-cylinder research engine has a production four-cylinder head, with only the #3 combustion chamber active. The laser beam enters from below, reflects off a mirror to pass horizontally through the exhaust stream, and then reflects off a second mirror to dump into the #4 exhaust port. The LII-RS signals are collected through a window in the top of the cell.

Preliminary Results

For convenience during this early development period, SNL is using a port fuel injected

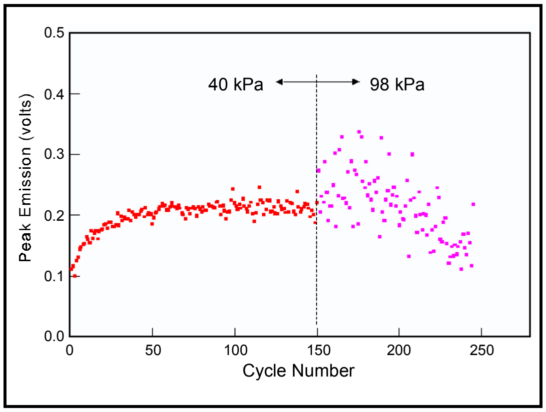

Figure 3. Preliminary LII measurements obtained during a snap-throttle test

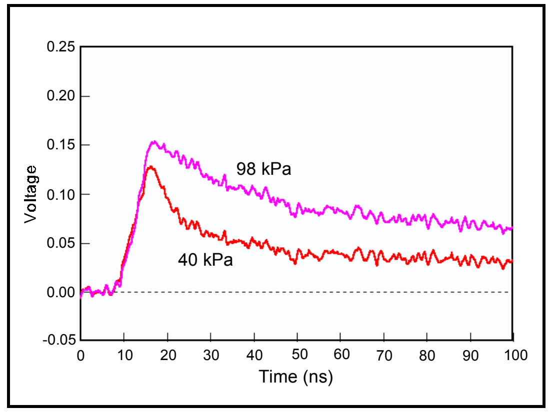

Figure 4. Temporal LII signals for before and after the snap throttle

gasoline engine rather than a diesel; during cold start, it will produce PM. Shown in Figure 3 are preliminary LII measurements obtained during a snap-throttle test for stoichiometric fueling at 1200 rpm. For the first 150 cycles the manifold pressure was 40 kPa, and then the throttle was snapped open to ambient (98 kPa) for 100 additional cycles. During the first 50 engine cycles the in-cylinder equivalence ratio increases as the engine warms and the port-wall fuel films develop, and this results in increased PM density. At the snap throttle the PM density dramatically increases, but also becomes very erratic. In addition, as the test progressed the LII signal intensity falls due to soiling of the optical cell windows. This is a problem that can be corrected by some redesign of the optical cell.

Finally, shown in Figure 4 are temporal LII signals for before and after the snap throttle. The faster decay rate for the throttled condition is indicative of smaller primary particle size, which is the expected trend.

List of Acronyms

LII	Laser Induced Incandescence
PM	Particulate Matter
RS	Rayleigh Scattering

V.B. Particulate Measurement Instrument Development

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Objectives

- Develop the Diesel Particle Scatterometer (DPS) for real-time diesel particle size measurements
- Use instrument to study particle characteristics as a function of: engine type; load, RPM; fuel

composition; and post-combustion processes (aftertreatment, dilution, etc.)
- Extend DPS capabilities in time, sensitivity, and application

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 1C, Barriers A,B

Accomplishments

- Designed, built, and calibrated two DPS instruments for real time particle size measurement
- Demonstrated operation at > 1 Hz data rates
- Installed and tested instruments at ORNL and LBNL
- Performed intercomparisons at ORNL with the Scanning Mobility Particle Sizer (SMPS) and the MOUDI Sampler
- Identified method for bi-modal particle size distribution analysis

Future Directions

- Continue DPS development to improve capabilities:
  - Incorporate bi-modal modeling capability into the DPS
  - Obtain a short wavelength (uv) laser
  - Add detector and software to monitor non-sphericity
  - Increase speed of instrument (software development)
  - Improve mechanical design (sealing, flow, and portability)
  - Develop diesel-like particle generator
  - Continue evaluation of commercial version of the DPS
- Expand dialogue with California regarding compliance ambient and air quality measurements

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Introduction

The viability of characterizing diesel particulates based on simultaneous fitting of three angle-dependent elements of the Mueller matrix for polarized light scattering has been established. Two instruments utilizing this approach and specialized for diesel particle detection have been designed, built and calibrated, see figure 1. One of these instruments has been installed in the engine

Figure 1. Diesel Particle Scatterometer

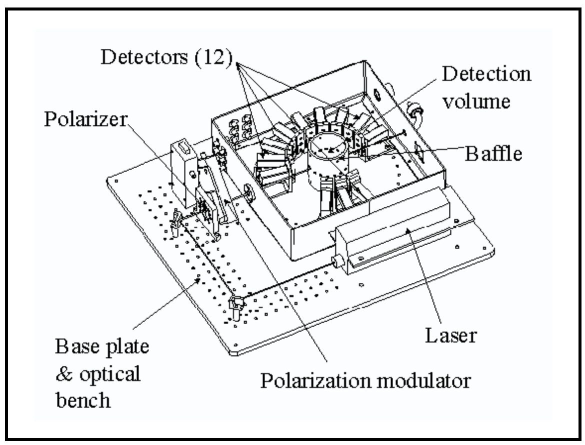

Figure 2. Diesel Particle Scatterometer Schematic

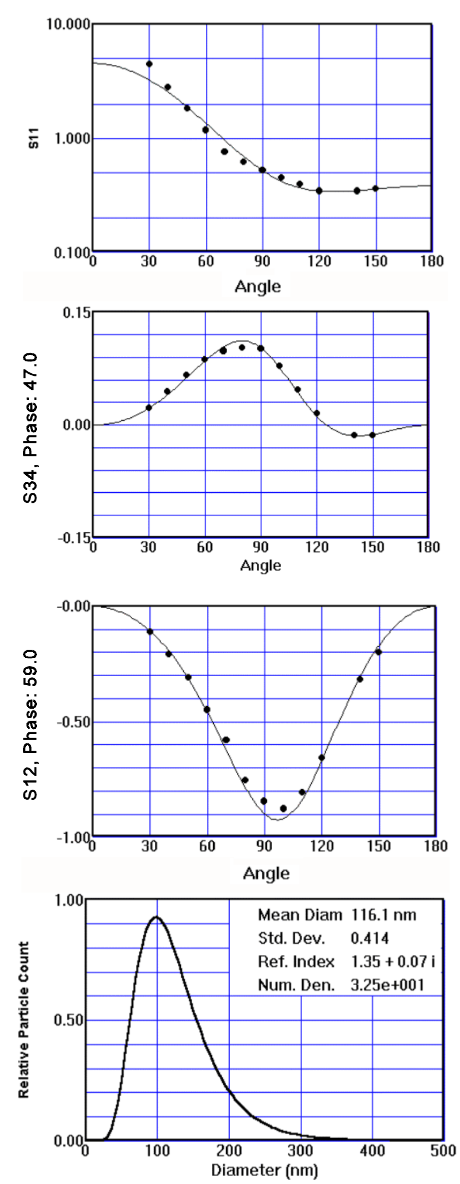

Figure 3. Matrix elements and resulting particle size distribution and optical properties

testing laboratory at Oak Ridge National Laboratory. Figure 2 is a schematic which illustrates the main components of the instrument.

The matrix elements are determined by the synchronous detection of the polarized laser light, modulated at 50 kHz by a polarization modulator, and by the total scattered light intensity. A portion of the diluted exhaust is passed through the measurement volume and the light scattered by the particulate stream is detected by an array of 12 compact programmable photomultiplier tubes (PMT). The measurement of the exhaust is therefore effectively performed in-situ.

The output from the PMTs is digitized and analyzed by lock-in detection with custom software developed at LBNL for this instrument. Thus, three matrix elements are obtained at 12 fixed angles and the output is displayed on a computer monitor, as shown in Figure 3. The results are then fitted with modeled scattering calculations using a Levenburg-Marquardt optimization technique to determine the particle size distribution and also refractive and absorptive optical properties of the particles. The absorptive component of the index of refraction provides a measure of the graphitic carbon content of the exhaust particles.

Figure 3 illustrates the three measured matrix elements and the calculated size distribution. These data were obtained from a Volkswagen TDI diesel engine running at 1900 rpm at full load and rated torque.

An important advantage of the instrument is the rapid response time; it has been tested at greater than 1 Hz data acquisition rate. This speed allows for the measurement of engine transients and even cylinder to cylinder variations. The data shown in Figure 3 were obtained at approximately a 2.5 Hz data rate.

Detailed intercomparisons of the DPS with a Scanning Mobility Particle Sizer (SMPS) have been carried out at Oak Ridge National Laboratory on the diluted exhaust from the TDI engine mentioned above. The SMPS is a slower instrument that scans through the particle sizes in ~2-3 minutes. Comparison of the particle size distributions obtained

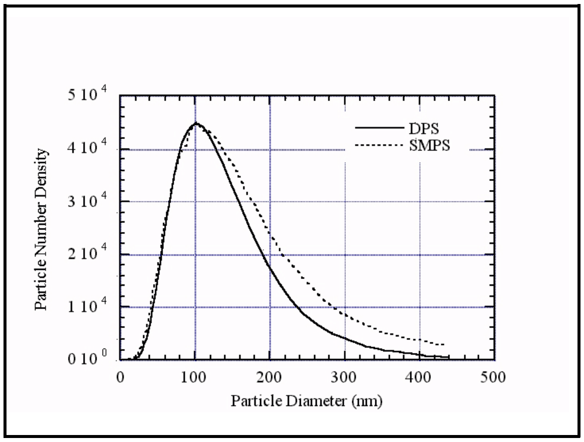

Figure 4. Comparison of results from the Diesel Particle Scatterometer and the Scanning Mobility Particle Sizer

simultaneously from the two instruments for the running condition illustrated above in Figure 3 are
shown in Figure 4. As can be seen the comparison is very good for this log-normal monomodal particle size distribution which has a mean particle diameter, determined by the DPS of 120 nm. At certain running conditions bi-modal particle distributions, due to the presence of large numbers of nanoparticles (< 10nm), were observed and strategies are being developed to include the affect of the light scattered by these very small particles in the analysis of the data.

Conclusions

Two instruments for rapid, in-situ measurements of diesel exhaust were built and calibrated to measure the size distribution and optical properties of a modern direct injected turbocharged engine. In initial testing, the DPS showed good sensitivity and discrimination of the diesel exhaust for a variety of running conditions and dilution ratios. As outlined above work will continue on improving the flexibility and sensitivity of the DPS instrument.

References/Publications

A.J. Hunt, M.S. Quinby-Hunt, I.G. Shepherd, "Diesel Exhaust Particle Characterization by Polarized Light Scattering," SAE Transactions 982629, 1998, LBNL-43695.

M.S. Quinby-Hunt, P.G. Hull, I.G. Shepherd, and A.J. Hunt, "Calculations of Light Scattering from Non-Spherical Aerosol Particles with Parallel Processing," Proc. Ocean Optics XIV, Kona, HI, Nov 10-14, 1998.

A.J. Hunt, M.S. Quinby-Hunt, I.G. Shepherd, "Polarized Light Scattering for Diesel Exhaust Particulate Characterization," Proc. of the Diesel Engine Emissions Reduction Workshop, in press 1998.

A.J. Hunt, I.G. Shepherd, M.S. Quinby-Hunt and J.M. Storey, "Size and Morphology Studies of Diesel Exhaust Particles using Polarized Light Scattering," 5th International Congress on Optical Particle Sizing, Minneapolis, MN Aug 10-13, 1998

A.J. Hunt, M.S. Quinby-Hunt, I.G. Shepherd, "Diesel Exhaust Particle Characterization by Polarized Light Scattering," Proc. Automotive Technology Development Coordination Meeting, Sept 27-30, Dearborn, MI. 1997.

List of Acronyms

DPS	Diesel Particle Scatterometer
LBNL	Lawrence Berkeley National Laboratory
MOUDI	Micro-Orifice Uniform Deposit Impactor
ORNL	Oak Ridge National Laboratory
PMT	Photomultiplier Tubes
RPM	Revolutions per minute
SMPS	Scanning Mobility Particle Sizer
TDI	Turbo Direct Injection

V.C. Ultrasonic Particulate Matter Monitor

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Objective

- Develop a low-cost, in-situ, and fast-response monitor for particulate matter (PM) to provide real-time control of diesel engine combustion.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 1C, Barriers A,B

Approach

- Conduct a feasibility study of applying ultrasonic techniques to monitoring of particulate matter.
- Develop a prototype monitor and evaluate its performance.
- Conduct field tests and transfer the technology to industry.

Accomplishments

- Established basic design of the ultrasonic PM monitor.
- Conducted laboratory tests to determine correlations between ultrasonic parameters and PM concentrations.

Future Directions

- Complete the feasibility study to establish the design, performance, and capabilities of the ultrasonic PM monitor.
- Design and test a prototype PM monitor.
- Conduct field tests and initiate technology transfer.

Introduction

For higher fuel efficiency and lower emissions, the U.S. Department of Energy (DOE), Office of Transportation Technologies (OTT), has launched an aggressive research and development (R&D) program to develop clean diesel engines that utilize compression-ignition direct-injection (CIDI) technology. CIDI diesel engines offer higher thermal efficiency than spark-ignited gasoline engines, but suffer from high emissions of NO_x and particulates.

Typically, submicrometer particles are generated in a combustion process. In the exhaust gas stream from a diesel engine, unburnt carbons and volatile matter, such as hydrocarbons and inorganic species are agglomerated to form these microscopic particles. Because of their long suspension time in air, the submicrometer particles are likely to cause more health concerns and thus need to be closely monitored. Emission of NO_x can be reduced by using lean-burn catalysts but this often results in increased PM emissions. Thus, better control of engine combustion is needed. For these reasons, one of the focus areas of CIDI engine research is development of low-cost, real-time emission sensors for NO_x and PM that provide real-time combustion control of CIDI diesel engines.

Optical techniques [1] have been widely employed for PM monitoring; measurements of light attenuation and scattering are generally used to determine particle concentration and size distribution, respectively. Typically, the particle-size distribution is determined from either the ratio of scattered light at a fixed angle for various incident light wavelengths [2] or the polarization ratio at various scattering angles for a fixed incident light wavelength [3]. Because of practical problems such as vibrational effect on the light source and surface contamination on optical windows, optical techniques are still limited to laboratory applications. Other commonly used PM measurement instruments [4] are impactors, electrical analyzers, photoelectric sensors, and time-of-flight mass spectrometers. However, these are basically laboratory instruments and are impractical for direct tailpipe application.

Ultrasonic techniques have been applied to measurements of particle size and concentration in solids suspension [5]. A commercially available instrument based on acoustic attenuation spectroscopy [6] and claims to measure particle concentration up to 50% by volume and size range of 0.01 to 1000 µm. Ultrasonic approaches have also been examined for use with emulsions, and more notably for water droplets in air. Theories on scattering and absorption have been thoroughly studied, but only limited measurement data have been reported, especially on aerosols and PM in air. The objective of this project is to develop ultrasonic techniques for PM monitoring.

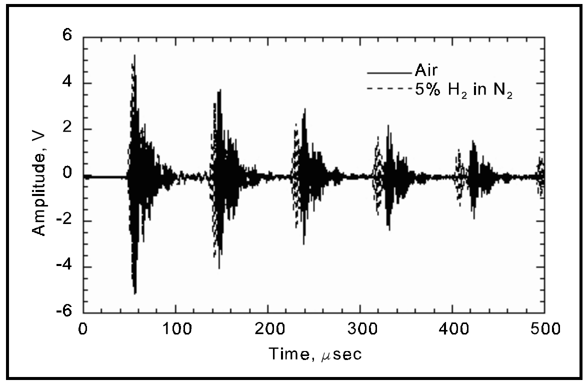

Figure 1. Ultrasonic Pulse Trains Detected in Two Gas Mixtures

Transducer and Measurement Principle

The basic design of the ultrasonic PM monitor consists of an acoustic cell, a high-frequency (0.5 MHz) piezoelectric transducer, and a flow pump. The transducer is designed to operate in gas media, generating ultrasonic pulses and receiving the pulses reflected from the cell wall. Because of the narrow cell design (1.27 cm), multiple reflections are observed. Figure 1 shows a typical reflection train detected from the transducer. Each consecutive reflection represents an additional 2.54 cm path length; thus the higher-order reflections provide higher sensitivity. As illustrated in Figure 1, the higher-order reflected peaks are better separated for different gas media. The acoustic technique, different from optical methods, measures two physical parameters: sound velocity and sound attenuation. Sound velocity in a gas, which can be measured from the

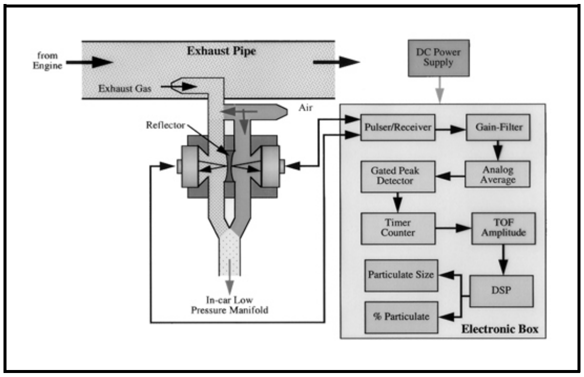

Figure 2. Conceptual Design of Ultrasonic Particulate Monitor

separation of reflections, can be calculated from:

$$C = \left(\frac{RT}{M} \; \frac{C_p}{C_p\text{-}R} \right)^{½} \quad (1)$$

where C is the sound speed, R the gas constant, M the molecular weight of the gas, and C_p the heat capacity under constant pressure. Equation 1 clearly shows that sound speed in a gas medium varies with the temperature, molecular weight and thermal properties of the gas. In principle, the presence of particulates has very little effect on sound speed, especially for low volume-percents of particulates, but the effect on attenuation is significant. In general, attenuation is caused by scattering, viscous effects due to the relative motion of the gas with respect to the particles, and thermal effects due to irreversible heat transfer between the gas and the particles. For submicrometer particles entrained in a gas stream at low temperature, scattering will be the dominant effect on attenuation. Because the wavelength of 0.5-MHz sound waves in air is about 670 µm (much greater than the particle size), the scattering cross section is proportional to the square of the particulate volume [7].

Figure 2 shows a conceptual design of the PM monitor with a dual-cell configuration that enables the sensor to make a relative measurement, thus minimizing environmental effects such as temperature and pressure fluctuations. Changes in sound speed and attenuation, measured in terms of time-of-flight (TOF) and integrated peak amplitude, respectively, will be measured and correlated with PM concentration, gas composition, and particle-size distribution.

Laboratory Test Results

A single-cell laboratory prototype was built for the feasibility study. A wide-band pulser/receiver (Panametrics model 5058PR)

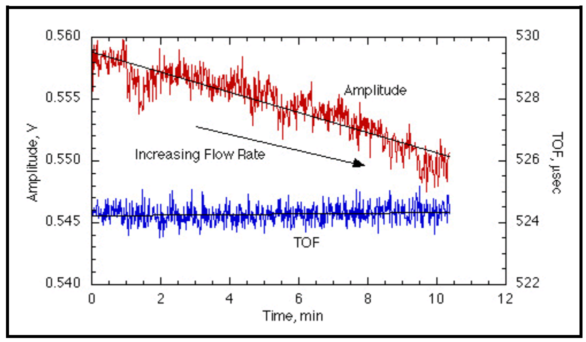

Figure 3. Effect of Flow Rate on Amplitude and TOF Measurements

was used to pulse the transducer and amplify the echoes by 40 dB. Echo output from the receiver passed through a bandpass filter (450 to 550 Hz) with another 20 dB gain. The fifth echo was then gated out and digitized for analysis of its TOF and amplitude. A flow control system and an aerosol generator (TSI model 3076) were connected to the prototype sensor.

Effects on signal characteristics due to changes of flow rate and gas temperature were examined for calibration sensor response. Figure 3 shows the amplitude and TOF variations over the range of the air-pump speed. Results indicate that TOF is not affected by the flow-rate change but that amplitude decreases as flow rate increases. To estimate the temperature effect, an aerosol

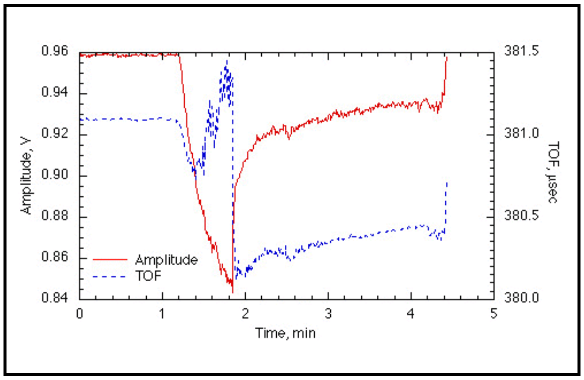

Figure 4. Temperature Effect on Amplitude and TOF Measurements

Table 1. Particle Diameters of Test Samples

Test Samples	Particle Size, μm (count media dia.)	Comments and References
NaCl aerosol	0.06	Based on TSI calibration data
Cigarette smoke	0.27	Reference 11
Joss stick smoke	0.13	Reference 12

conditioner (TSI model 3072) was used. The result is shown in Figure 4. Generally, both TOF and amplitude vary with temperature; a higher temperature results in lower TOF (higher sound speed) and amplitude. Both changes are as predicted by theory.

To demonstrate the technique and estimate the sensitivity, three types of test samples were used: sodium chloride aerosol, cigarette smoke, and joss stick smoke. Table 1 lists the estimated particle sizes of the test samples. The NaCl aerosol was generated from a 5% NaCl solution, cigarette smoke was from a Marlboro Light cigarette, and the joss stick was 1.6 mm in diameter. Figure 5 shows TOF and amplitude changes for the aerosol test.

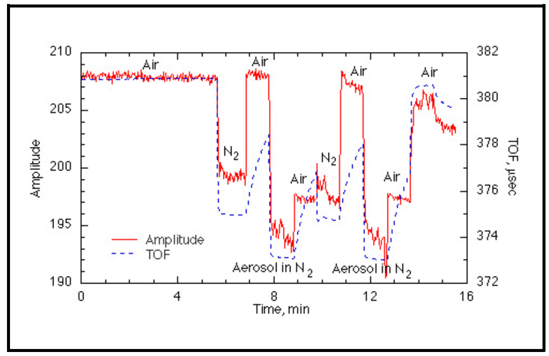

Figure 5. Amplitude and TOF Changes in NaCl Aerosols

The presence of NaCl aerosol in the carrier gas (dry nitrogen gas) decreases both TOF and amplitude. The decrease in TOF, which means an increase in sound speed, implies that either the aerosol temperature is higher or the molecular weight of the aerosol flow is lower than those of nitrogen gas. The latter is more likely due to entrained moisture.

Figure 6 shows the results of the cigarette test. An approximate 30% drop in signal amplitude was measured, but the TOF increased by 10 μm. The decrease in sound speed may be due to gases of greater molecular weight, for example CO_2. Similar trends, as shown in Figures 7 and 8, were observed for joss-stick smoke. Filters were used in the joss-stick tests to collect particulates. Figure 9 shows the amplitude change as a function of mass flow rate of the joss-stick smoke. A linear dependence can be obtained, giving an

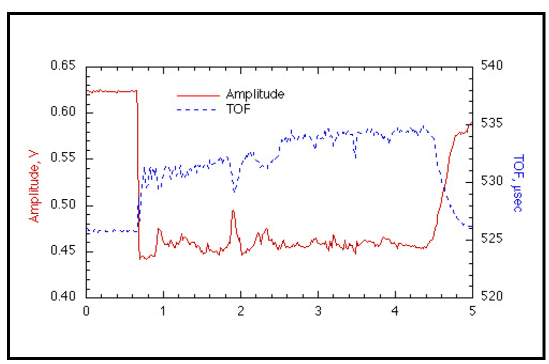

Figure 6. Amplitude and TOF Changes in Cigarette Smoke

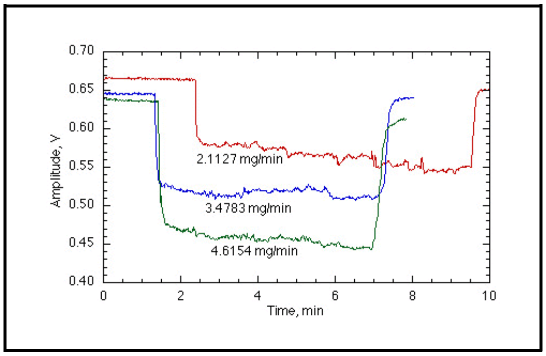

Figure 7. Amplitude Changes in Joss-stick Smoke of Different Mass Flow Rates

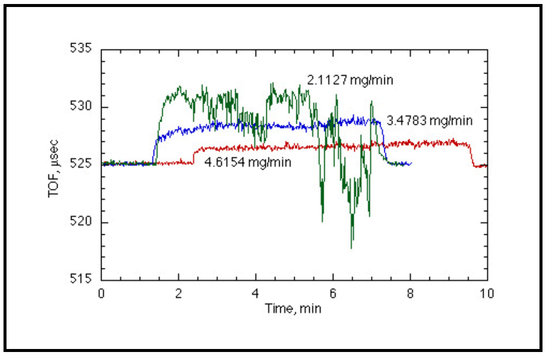

Figure 8. TOF Change in Joss-stick Smoke of Different Mass Flow Rates

estimated sensitivity of 0.4 μg/sec-mv.

Conclusions and Future Plans

The feasibility of using ultrasound to monitor PM concentration was studied. A laboratory prototype sensor was built and tested with aerosols and smoke. Results show that both sound speed and attenuation change with PM concentration, but that sound speed is more sensitive to gas-phase chemical composition, while attenuation shows a more direct dependence on PM concentration. Effects on sensor response due to changes in gas temperature, pressure, and flow rate were also examined. Flow rate affects attenuation measurement only slightly, but both temperature and pressure showed significant effects on both sound speed and attenuation. Thus, a dual-cell design with one cell used for reference is desirable.

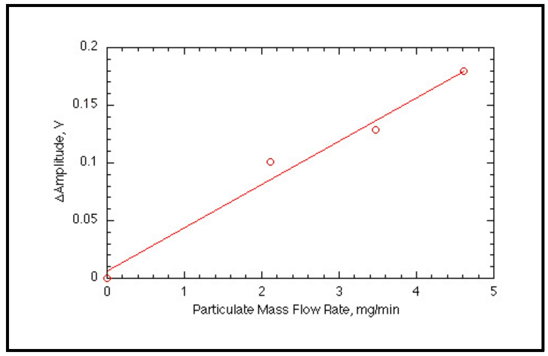

Figure 9. Amplitude Change vs. Particulate Mass Flow Rates

Development of the ultrasonic PM sensor is required in order to conduct more tests to determine sensor performance. The tests will include different types of aerosols and particulates. Detection of particle-size distribution will also be examined. A prototype sensor with dual-cell configuration will be built and tested. Required control electronics and software will be developed.

References

1. H. C. van de Hulst, "Light scattering by small particles," John Wiley & Sons, Inc. (1957).
2. A. F. Stevenson, W. Heller, and M. L. Wallach, J. Phys. Chem. V. 34, pp. 1789-1795 (1961).
3. R. T. Jacobsen, M. Kerker, and E. Matijevic, J. Phys. Chem. V. 71, p. 514 (1967).
4. D. B. Kittelson, "Engines and nanoparticles: A review," J. Aerosol Sci., V. 29 (5/6), pp. 575-588 (1998).
5. P. S. Epstein and R. R. Carhart, "The absorption of sound in suspensions and emulsions, I. Water fog in air," J. Acoustic Soc. of Am. V. 25 (3), pp. 553-565 (1953).
6. "Ultrasizer," Malvern Instruments Ltd., Malvern, UK.
7. A. Ishimaru, "Wave propagation and scattering in random media," V. 1, p. 13, Academic Press (1978).

List of Acronyms

CIDI	Compression Ignition Direct Injection
OTT	Office of Transportation Technologies
PM	Particulate Matter
TOF	Time-of-Flight

V.D. Real-time Particulate Monitoring for Emissions Control

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Objectives

- Use a recently developed advanced laser based diagnostic for particulate emission measurement in diesel exhausts.
- Assess implications towards the feasibility of a portable instrument for real-time particulate measurement. Signals from such an instrument could be used for closed-loop control for emissions reduction.

Office of Advanced Automotive Technologies R&D Plan, Advanced Heat Engines: Task 1C, Barriers A,B

Approach

- Validate the use of Laser Induced Incandescence (LII) for the measurement of particulate mass concentration, M (g/cc), in diesel exhausts.
- Verify the feasibility of additionally measuring particulate number density, N (particulates/cc) and mean size, D (nm), by simultaneous execution of LII with Laser Light Scattering (LLS).
- Also, verify determination of the fraction of Volatile Organic Compounds (VOC) through Laser Induced Fluorescence (LIF).

Accomplishments

- Facilities required for conducting above experiments have been set up.
- Designed and fabricated a laminar diffusion burner to serve as a calibration standard for LII signals.
- Fabricated and tested physical sampling systems for LII technique validation. These constitute a mini-dilution tunnel, Condensation Particulate Counter, and a TEOM instrument.

Future Directions

- Perform particulate measurements in the exhausts of lab generated flames.

- Conduct experiments using LII and associated techniques for particulate characterization in diesel exhausts, both under steady state as well as transient conditions.
- Issues related to the development of a stand-alone / add-on unit for particulate measurements in engine exhausts to be assessed.

Introduction

With anticipated future particulate emission regulations to be based on number density, N (particles/cc), in addition to the traditional mass concentration, M (gms/cc), there is an impending need for advanced diagnostics for particulate measurement. The current EPA approved technique for diesel exhausts entails the collection of particulates over a filter in a diluted stream of exhaust gases which is followed by gravimetric analysis. Such a technique is limited to measurements at steady state operation while requiring appreciable operator skill and time. Also, particulate emission characterizations during transient cycles as prescribed in the Federal Test Procedures cannot be performed using such a technique. Other traditional techniques using CPC or TEOM are limited in their capabilities and suffer from long response times.

On the other hand, Laser Induced Incandescence (LII), a recently developed technique, allows highly time resolved (~0.05 s resolution) planar imaging of soot concentrations in diesel exhausts without any need for dilution. Also, LII can be used for measurements during the transient cycle evaluations as prescribed in the Federal Test Procedures. Furthermore, it can be combined with other laser based techniques to obtain information such as particle number concentration, N (particles/cc) and size, (D nm), in addition to the fraction of volatile organic compounds (VOC) that accompany soot emissions. It is the objective of the present study to validate the application of LII and other associated techniques for particulate measurements in diesel exhausts.

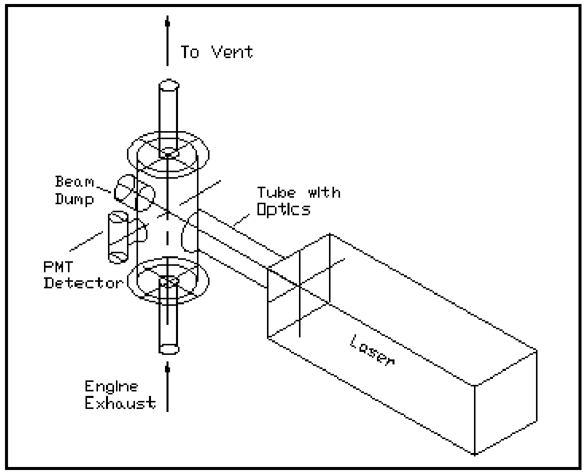

Figure 1. Experimental setup for implementing I-D LII. For planar measurements, the PMT detector will be replaced with an Intensified CCD camera.

Accomplishments

So far, efforts have been pursued in acquiring all the elements required for the experimental setup (cf. Figure 1.). Also, instrumentation required for technique validation has been established. Further details are provided below.

A laser with the required power levels has been refurbished and is readily available. The required instrumentation for 2-D imaging - Intensified gated CCD camera & the Nikon lens - have been purchased. Also, instrumentation for performing point measurements - Photo Multiplier Tube, high voltage power supply & lenses - have been obtained. Concurrent to these efforts, a laminar diffusion flame burner (cf. Figure 2.) was designed and fabricated. The stable flame from this burner will be used for calibration of the LII signal. A laboratory to accommodate

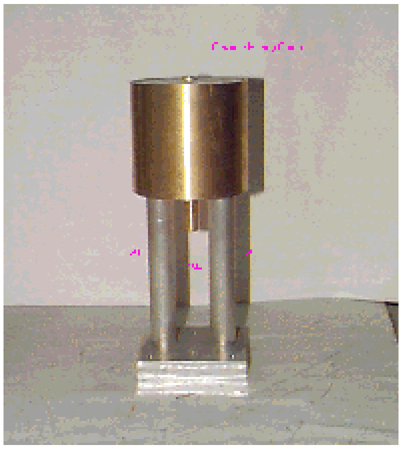

Figure 2. A Panoramic View of the Laminar Diffusion Flame Burner

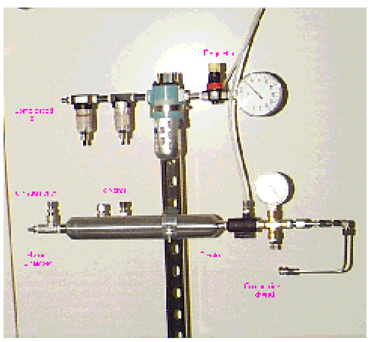

Figure 3. University of Minnesota Single Stage Micro Dilution Tunnel for Use with Diesel Exhaust

the above facility is currently being setup and will be operational in late July, 1999.

Meanwhile, efforts have been focused in preparing instrumentation for validation of the LII technique. A single stage dilution tunnel was fabricated based on a design developed by Prof. Kittleson at the University of Minnesota (cf. Figure 3). This dilution tunnel while being compact is based on the ejector concept. By varying the air pressure and the orifice size, the dilution ratio can be varied between 5 and 35. Calibration curves for achievable dilution ratios were estimated and are presented below (cf. Figure 4). Preliminary measurements of

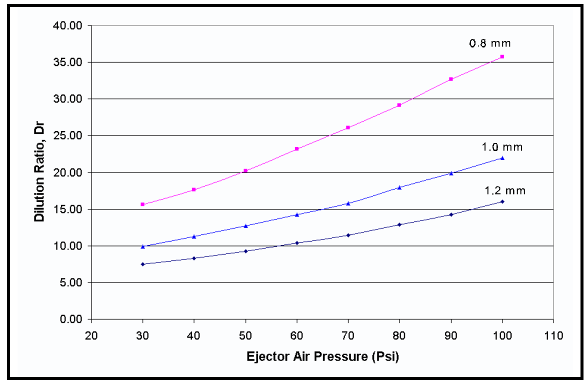

Figure 4. Achievable dilution ratios in the single stage dilution tunnel. Curves for three different orifices are shown.

particulate emissions in the exhaust of a turbo-charged VW engine were performed. The diluted exhaust sample was passed through Tapered Element Oscillating Microbalance (TEOM) & Condensation Particulate Counter (CPC) instruments to obtain characteristics of the particulate emissions. Typical TEOM traces using this system for measurements in a diesel exhaust are given in Figure 5. This system will be used for validation of LII measurements during engine steady state operations.

Future Plans

Following commissioning of the facility in late July, the following activities are planned in the immediate future:

1) Implement 1-D & 2-D LII in a standard flame for calibration purposes.
2) Perform measurements in the exhaust stream of lab generated flames for validation purposes.
3) If possible perform preliminary measurements in obtaining the fraction of Volatile Organic Compounds (VOC).

Following such efforts, the LII&LIF and LII&LLS techniques will be performed on the exhaust of a single cylinder Caterpillar engine operating in steady state conditions. Such

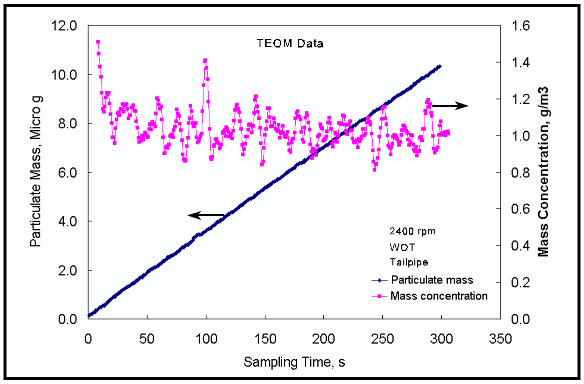

Figure 5. Traces of total mass deposited on the micro-filter (g) and mass concentration (g/m^3) in a typical diesel exhaust obtained using a single stage dilution tunnel and a TEOM

measurements will be validated with those performed using standard particulate sampling procedures. Subsequently, the applicability of the above techniques for transient cycles will also be tested. It is planned to use the recently established advanced power train test facility of ANL, which facilitates measurements during the transient test cycles.

Finally, implications will be drawn towards the development of a portable LII based instrument for diesel exhaust measurement. Depending upon the available resources, efforts will be pursued in the development of a prototype.

Publications/Presentations

S. Gupta, Patent filed for "A Portable LII Based Instrument for Diesel Exhaust Particulate Measurement."

List of Acronyms

ANL	Argonne National Laboratory
CCD	Charge-Coupled Device
CPC	Condensation Particulate Counter
LIF	Laser Induced Fluorescence
LII	Laser Induced Incandenscence
LLS	Laser Light Scattering
TEOM	Tapered Element Oscillating Microbalance
VOC	Volatile Organic Compounds